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Kinetic energy estimates for the accuracy of the time-dependent Hartree–Fock approximation with Coulomb interaction

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ABSTRACT

We study the time evolution of a system of N spinless fermions in $\mathbb{R}^3$ which interact through a pair potential, e.g., the Coulomb potential. We compare the dynamics given by the solution to Schrödinger's equation with the time-dependent Hartree–Fock approximation, and we give an estimate for the accuracy of this approximation in terms of the kinetic energy of the system. This leads, in turn, to bounds in terms of the initial total energy of the system.

RÉSUMÉ

On étudie l'évolution temporelle d'un système de N fermions sans spin dans $\mathbb{R}^3$ qui interagissent via un potentiel à deux particules, par exemple, le potentiel de Coulomb. On compare la solution de l'équation de Schrödinger avec son approximation donnée par la méthode de Hartree–Fock dépendant du temps, et on estime la précision de cette approximation en fonction de l'énergie cinétique du système. De ceci découle une borne de l'erreur en fonction de l'énergie totale du système.

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1. Introduction

The model. In quantum mechanics, the state of a system of N identical particles is described by a wave function Ψ_t which evolves in time $t \in \mathbb{R}$ according to Schrödinger's equation,

$$\begin{cases} i\partial_t \Psi_t = H\Psi_t \,, \\ \Psi_{t=0} = \Psi_0 \,. \end{cases} \tag{1}$$

Given the (Bose–Einstein or Fermi–Dirac) particle statistics and the one-particle Hilbert space $\mathfrak{h}$, the wave function Ψ_t is a normalized vector in $\mathfrak{H}_b^{(N)} := \mathcal{S}^{(N)}[\mathfrak{h}^{\otimes N}]$, for a system of N bosons, or in $\mathfrak{H}_f^{(N)} := \mathcal{A}^{(N)}[\mathfrak{h}^{\otimes N}]$, for a system of N fermions. Here $\mathcal{S}^{(N)}$ and $\mathcal{A}^{(N)}$ are the orthogonal projections onto the totally symmetric and the totally antisymmetric subspace, respectively, of the N-fold tensor product $\mathfrak{h}^{\otimes N}$ of the one-particle Hilbert space $\mathfrak{h}$. The dynamics (1) is generated by the Hamilton operator H which is self-adjointly realized on a suitable dense domain in $\mathfrak{H}_b^{(N)}$ or $\mathfrak{H}_f^{(N)}$, respectively.

In the present article we study a system of N spinless fermions in $\mathbb{R}^3$, so $\Psi_t \in \mathfrak{H}_f^{(N)}$, and $\mathfrak{h} = L^2[\mathbb{R}^3]$ is the space of square-integrable functions on $\mathbb{R}^3$. The Hamiltonian is given by

$$H \;=\; \nu + \sum_{j=1}^{N} h_j^{(1)} + \lambda \sum_{1\le j<k\le N} v(x_j - x_k) \,, \tag{2}$$

where

- the number $\nu \in \mathbb{R}$ is a constant contribution to the total energy. For example, if we describe a molecule in the Born–Oppenheimer approximation, then ν would account for the nuclear–nuclear repulsion,
- the coupling constant $\lambda > 0$ is a small parameter and possibly depends on the particle number $N \geq 1$ (while our interest ultimately lies in the description of systems with $N \gg 1$, the estimates in this article hold for any $N \geq 1$),
- the self-adjoint operator $h^{(1)}$ on $\mathfrak{h}$ is of the form $-a\Delta + w(x)$, where $a > 0$ and the external potential w is an infinitesimal perturbation of the Laplacian,
- and $v(x) := \pm|x|^{-1}$ is the Coulomb potential, for $x \in \mathbb{R}^3 \setminus \{0\}$; $v(x) = +|x|^{-1}$ is the repulsive case, $v(x) = -|x|^{-1}$ the attractive case.

The Hamiltonian specified in (2) describes several situations of interest, e.g.:

- **Atom.** For an atom in the (0^{th}) Born–Oppenheimer approximation with a nucleus of charge Z at the origin, we have repulsive interaction and

$$\nu = 0 \,, \quad h^{(1)} = -\frac{\Delta}{2} - \alpha\frac{Z}{|x|} \,, \quad \lambda = \alpha \,, \tag{3}$$

  where $\alpha > 0$ is the fine structure constant whose physical value is $\alpha \simeq 1/137$. Note that our system of units is chosen such that the reduced Planck constant $\hbar$, the electron mass m and the speed of light c are equal to one, and the charge of the electron is $-e = -\sqrt{\alpha}$. For more details about this choice of units see [45, p. 21].
- **Molecule.** More generally, we can consider a molecule with $M \in \mathbb{N}$ nuclei of charges $Z_1, \ldots, Z_M > 0$ at fixed, distinct positions $R_1, \ldots, R_M \in \mathbb{R}^3$ in the Born–Oppenheimer approximation. In this case we have

$$\nu = \sum_{1\le m<l\le M} \frac{\alpha Z_m Z_l}{|R_m - R_l|} \,, \quad h^{(1)} = -\frac{\Delta}{2} - \sum_{m=1}^{M} \frac{\alpha Z_m}{|x - R_m|} \,, \quad \lambda = \alpha \,. \tag{4}$$

- **Particles in a trap.** For electrons in an external confining potential (realized, e.g., by a laser trap), we have repulsive interaction and

$$\nu = 0\,, \quad h^{(1)} = -\frac{\Delta}{2} + w(x)\,, \quad \lambda = \alpha\,. \tag{5}$$

- **Fermion star.** The Hamiltonian also describes systems of gravitating fermions, e.g., neutrons. In this case the interaction is attractive and

$$\nu = 0\,, \quad h^{(1)} = -\frac{\Delta}{2}\,, \quad \lambda = G\,, \tag{6}$$

where G is Newton's gravitational constant (and recall that we set the mass $m = 1$). A better description of a fermion star is achieved by replacing the non-relativistic Laplacian by the semi-relativistic operator $\sqrt{-\Delta + \mathbb{1}}$.

For these situations the Hartree–Fock description that we are aiming at in this article and that we describe below can only be expected to hold for very short times (short relative to the large particle number N). For times of order 1, we have to choose the coupling constant small in N to see Hartree–Fock behavior ("mean-field scaling"). There are several possibilities to do that

- **Mean-field scaling for large volume.** Let us first note that for systems with large volume proportional to N, the kinetic energy is naturally also of order N. For such a system, the choice

$$\nu = 0, \quad h^{(1)} = -\frac{\Delta}{2} + w(x)\,, \quad \lambda = \frac{1}{N^{2/3}} \tag{7}$$

leads to an interaction energy which is of the same order in N as the kinetic energy (see [50] for a more detailed discussion).
- **Mean-field scaling for fixed volume.** For systems with volume independent of N, the mean-field limit is naturally coupled to a semi-classical limit. Note that here the kinetic energy is of order $N^{5/3}$. Then the choice

$$\nu = 0, \quad h^{(1)} = -\frac{\Delta}{2N^{1/3}} + w(x)\,, \quad \lambda = \frac{1}{N^{2/3}} \tag{8}$$

leads to an interaction energy of the same order as the kinetic energy and nontrivial mean-field behavior (see in particular [22,16] for more details).
- $\boldsymbol{\lambda = N^{-1}}$ **scaling.** Very often, the term "mean-field scaling" is identified with the choice $\lambda = N^{-1}$. However, comparing with (7) and (8), in the two situations considered above, we see that this scaling leads to a subleading interaction.

Theory of the time-dependent Hartree–Fock equation. Although (1) admits the explicit solution $\Psi_t = e^{-itH}\Psi_0$, this explicit form is not useful in practice (from the point of view of numerics, for example) because of the large number $N \gg 1$ of variables, and it therefore becomes necessary to consider approximations to this equation. One such approximation consists of restricting the wave function Ψ_t to a special class of wave functions. For fermion systems, the Hartree–Fock approximation is a natural choice: it restricts Ψ_t to the class of Slater determinants, i.e., to those $\Phi \in \mathfrak{H}_f^{(N)}$ which assume a determinantal form,

$$\Phi(x_1, \dots, x_N) = \frac{1}{\sqrt{N!}} \det \begin{pmatrix} \varphi_1(x_1) & \cdots & \varphi_1(x_N) \\ \vdots & \ddots & \vdots \\ \varphi_N(x_1) & \cdots & \varphi_N(x_N) \end{pmatrix}\,, \tag{9}$$

where the orbitals $\varphi_1, \dots, \varphi_N \in \mathfrak{h}$ are orthonormal. We express (9) more concisely as $\Phi = \varphi_1 \wedge \cdots \wedge \varphi_N$. In time-independent Hartree–Fock theory, one is interested in determining the minimal energy expectation when varying solely over Slater determinants [7,11,10,43,9], i.e., one is interested in finding

$$\inf \left\{ \langle \Phi, H\Phi \rangle \,\middle|\, \Phi = \varphi_1 \wedge \cdots \wedge \varphi_N \,, \quad \langle \varphi_i, \varphi_j \rangle = \delta_{ij} \right\}.$$

One can also study the evolution governed by (1) using Slater determinants, which gives rise to time-dependent Hartree–Fock theory. Here the basic intuition is that, for a system containing a large number of particles, the solution will stay close to a Slater determinant (at least for short times), provided the initial state is close to a Slater determinant. Turning this intuition into mathematics requires the specification of the equation of motion of the approximating Slater determinant, as well as a mathematically rigorous notion of being "close". For the derivation of the former, one assumes that the solution to (1) is of the form $\Phi_t = \varphi_{t,1} \wedge \cdots \wedge \varphi_{t,N}$, as in (9). It is then easy to verify that the orbitals $\varphi_{t,1}, \dots, \varphi_{t,N}$ necessarily satisfy the time-dependent Hartree–Fock (TDHF) equation, that is the system of N non-linear equations given by

$$i\frac{d\varphi_{t,j}}{dt} \;=\; h^{(1)}\varphi_{t,j} + \lambda \sum_{k=1}^{N} \Big([v * |\varphi_{t,k}|^2]\varphi_{t,j} - [v * (\varphi_{t,j}\bar{\varphi}_{t,k})]\varphi_{t,k} \Big) \tag{10}$$

for $j = 1, \dots, N$ ($\bar{\varphi}$ is the complex conjugate of φ).

The TDHF equation (10) can be rewritten in terms of the one-particle density matrix $p_t = \sum_{j=1}^{N} |\varphi_{t,j}\rangle\langle\varphi_{t,j}|$ with $\varphi_{t,j} \in \mathfrak{h}$ and $\langle \varphi_{t,j}, \varphi_{t,k} \rangle = \delta_{jk}$ as

$$\text{(TDHF)} \qquad i\partial_t p_t \;=\; [h^{(1)}, p_t] + \lambda \operatorname{Tr}_2[v^{(2)}, (p_t \otimes p_t)(\mathbb{1} - \mathfrak{X})]\,. \tag{11}$$

Here $\mathfrak{X}$ is the linear operator on $\mathfrak{h} \otimes \mathfrak{h}$ such that $\mathfrak{X}(\varphi \otimes \psi) = \psi \otimes \varphi$ and Tr_2 is the partial trace (see (23)). Sometimes, we write $p_t^{(2)} = (p_t \otimes p_t)(\mathbb{1} - \mathfrak{X})$. In the sequel, when speaking of the TDHF equation, we refer to (11). The term involving $\mathbb{1}$ is called the *direct term*, the term involving $\mathfrak{X}$ the *exchange term*.

Note that the TDHF equation (11) can be written as $i\partial_t p_t = [h^{(1)}_{HF}(p_t), p_t]$, where the effective HF-Hamiltonian $h^{(1)}_{HF}(\gamma)$ is given by

$$h^{(1)}_{HF}(\gamma) \;:=\; h^{(1)} + \lambda \operatorname{Tr}_2[v^{(2)}(\mathbb{1}_{\mathfrak{h}\otimes\mathfrak{h}} - \mathfrak{X})(\mathbb{1}_{\mathfrak{h}} \otimes \gamma)]\,. \tag{12}$$

Implicitly assuming the existence and regularity of p_t, the HF-Hamiltonian $h^{(1)}_{HF}(p_t)$ is self-adjoint with the same domain as $h^{(1)}$, and hence the solution to $\partial_t U_{HF,t} = -ih^{(1)}_{HF}(p_t)U_{HF,t}$, with $U_{HF,0} = \mathbb{1}$, is unitary. This has the important consequence that (11) preserves the property of the one-particle density matrix p_t of being a rank-N orthonormal projection. In other words, if $\Phi_t \in \mathfrak{H}_f^{(N)}$ evolves according to the TDHF equation and $\Phi_0 = \varphi_1 \wedge \cdots \wedge \varphi_N$ is a Slater determinant, then so is Φ_t, for all $t \in \mathbb{R}$.

The TDHF equation for density matrices as in (11) has been studied in [17] for a bounded two-body interaction. Then the mild solutions of the TDHF equation in the form (10) have been handled for a Coulomb two-body potential in [21] for initial data in the Sobolev space H^1. This result has been extended to the TDHF equation in the form (11) in [18,20]. Note that [18] also handles the case of a more general class of two-body potentials and the existence of a classical solution for initial data in a space similar to the Sobolev H^2 space for density matrices. In [63] the existence of mild solutions of the TDHF in the form (10) was proved for a Coulomb two-body potential with an (infinite sequence of) initial data in L^2. For the convenience of the reader we state the precise results we use about the theory of the TDHF equation in Appendix A. In [5] the existence and uniqueness of strong solutions to the von Neumann–Poisson equation, another nonlinear self-consistent time-evolution equation on density matrices, are proved with the use of a generalization of the Lieb–Thirring inequality. Another direction in which to generalize the Hartree equations is to consider,

instead of an exchange term, a dissipative term in the Hartree equation; the existence and uniqueness of a solution for such an equation have been proved in [6].

One-particle density matrix. The notion of proximity of two states we use in this article is defined by expectation values of k-particle observables, where $1 \leq k \ll N$. More specifically, if $\Psi_t \in \mathfrak{H}_f^{(N)}$ is the (normalized) solution to (1) and $\Phi_{HF,t} = \varphi_{t,1} \wedge \cdots \wedge \varphi_{t,N}$, where $\varphi_{t,1}, \ldots, \varphi_{t,N}$ are the solutions to (10), then, for any k-particle operator $A^{(k)}$ (i.e., for any bounded operator $A^{(k)}$ on $\mathfrak{h}^{\wedge k} := \mathcal{A}[\mathfrak{h}^{\otimes k}]$), we wish to control the quantity

$$\delta_t^{(k)}\big(A^{(k)}\big) \;:=\; \frac{1}{\|A^{(k)}\|_\infty}\big|\langle \Psi_t,\ (A^{(k)} \otimes \mathbb{1}_{N-k})\Psi_t\rangle - \langle \Phi_{HF,t},\ (A^{(k)} \otimes \mathbb{1}_{N-k})\Phi_{HF,t}\rangle\big|\,.$$

Here $\mathbb{1}_{N-k}$ denotes the identity operator on $\mathfrak{h}^{\otimes(N-k)}$ and $\|\cdot\|_\infty$ denotes the operator norm on $\mathcal{B}[\mathfrak{h}^{\wedge k}]$.

It is more convenient to reformulate this approach in terms of reduced density matrices. We recall that, given $\Psi \in \mathfrak{H}_f^{(N)}$, the corresponding reduced k-particle density matrix is the trace-class operator $\gamma_\Psi^{(k)}$ on $\mathfrak{H}_f^{(k)}$ whose kernel is given by

$$\begin{aligned}&\gamma_\Psi^{(k)}(x_1, \ldots, x_k; y_1, \ldots y_k)\\ &\quad= \frac{N!}{(N-k)!}\int \Psi(x_1, \ldots x_k, x_{k+1}, \ldots x_N)\,\overline{\Psi(y_1, \ldots y_k, x_{k+1}, \ldots x_N)}\; d^3x_{k+1}\cdots d^3x_N\,.\end{aligned} \tag{13}$$

Note that we normalize the reduced density matrices so that $\operatorname{Tr}\gamma_\Psi^{(k)} = \frac{N!}{(N-k)!}$. We may then rewrite $\delta_t^{(k)}(A^{(k)})$ as

$$\delta_t^{(k)}\big(A^{(k)}\big) = \frac{1}{\|A^{(k)}\|_{\mathcal{B}(\mathfrak{H}_f^{(k)})}}\Big|\operatorname{Tr}\big[(\gamma_{\Psi_t}^{(k)} - \gamma_{\Phi_{HF,t}}^{(k)})A^{(k)}\big]\Big|$$

and observe that

$$\sup_{A^{(k)} \in \mathcal{B}(\mathfrak{H}_f^{(k)})} \delta_t^{(k)}\big(A^{(k)}\big) = \big\|\gamma_{\Psi_t}^{(k)} - \gamma_{\Phi_{HF,t}}^{(k)}\big\|_{\mathcal{L}^1}\,,$$

where $\|\cdot\|_{\mathcal{L}^1}$ denotes the trace norm. We are thus interested in bounds on $\|\gamma_{\Psi_t}^{(k)} - \gamma_{\Phi_{HF,t}}^{(k)}\|_{\mathcal{L}^1}$. In the present article we restrict ourselves to the case $k = 1$.

Derivation of the TDHF equation. The derivation of the TDHF equation may be seen as part of the quest for a derivation of macroscopic, or mesoscopic, dynamics from the microscopic classical or quantum-mechanical dynamics of many-particle systems as an effective theory. Let us first discuss some generally interesting examples and then come to the case of the TDHF equation for fermions.

In the case of the dynamics of N identical quantum-mechanical particles, the time-dependent Hartree equation, that is the TDHF equation (10) without the exchange term, was first derived rigorously in [60] for a system of N distinguishable particles in the mean-field limit. For systems of indistinguishable particles, the case of bosons has received considerable attention compared to the case of fermions, and several methods have been developed. The so-called Hepp method has been developed in [41,35,36] in order to study the classical limit of quantum mechanics. It inspired, among others, [34], where the convergence to the Hartree equation is proved, [57], where the rate of convergence toward mean-field dynamics is studied, and [2,3], where the propagation of Wigner measures in the mean-field limit is studied, with special attention to the relationships with microlocal and semi-classical analysis. In this direction, with a stochastic microscopic model, the linear Boltzmann equation was obtained as a weak-coupling limit in [19] yielding an example for a derivation of an equation with non-local terms using methods of pseudodifferential calculus. The derivation

of the linear Boltzmann equation in the earlier work [30], along with the series of works following it, used a different method based on series expansions in terms of graphs similar to Feynman diagrams. The result is valid on longer time-scales than in [19], but with more restrictive initial data. Other limit dynamics have been obtained, a particularly interesting one is the weak-coupling limit for interacting fermions for which a (non-rigorous) derivation of the nonlinear Boltzmann equation has been given in [25]. Series expansion methods and the Bogoliubov–Born–Green–Kirkwood–Yvon (BBGKY) hierarchy have also proved fruitful in other works, e.g., [60,14,23,1,27,4,29,28]. In [29,28] the Gross–Pitaevskii equation, which describes the dynamics of a Bose–Einstein condensate has been derived. Also for the Gross–Pitaevskii equation the formation of correlations has been studied in [24], providing information on the structure of solutions to the Gross–Pitaevskii equation. The techniques developed in [48] to study the weakly nonlinear Schrödinger equation are used in [47] to derive quantum kinetic equations; those techniques resemble the BBGKY hierarchy methods, but they do not impose the normal ordered product of operators when considering expectation with respect to the initial state. The bounds on the rate of convergence in the mean-field limit given in [34] have been sharpened in [26] using a method inspired by Lieb–Robinson inequalities. Another method introduced in [33] shows that the classical time evolution of observables commutes with the Wick quantization up to an error term which vanishes in the mean-field limit, yielding an Egorov-type theorem. Recently a new method based on a Grönwall lemma for a well-chosen quantity has been introduced [52,42] in the bosonic case, which considerably simplifies the convergence proof for the Hartree equation.

In the fermionic case, the TDHF equation has been derived in [12] in the $\lambda = N^{-1}$ scaling for initial data close to Slater determinants, and with bounded two-body potentials. The same authors give bounds on the accuracy of the TDHF approximation for uncorrelated initial states in [13], still with a bounded two-body potential. For the same scaling, the TDHF equation has been derived in [32] for the Coulomb potential for sequences of initial states given by Slater determinants. The semi-classical mean-field scaling from (8) has first been considered in [49] where it is shown that for suitably regular interactions the Schrödinger dynamics is close to the classical Vlasov dynamics. The results have been improved in [61]. In [22], in the semi-classical mean-field scaling, the closeness of the Schrödinger dynamics to the Hartree–Fock dynamics was discussed and bounds for the Husimi function were given, assuming the potential to be real-analytic and thus in particular bounded. Up to that point all the method used to derive the TDHF equation had always been based on BBGKY hierarchies. In [16,15] estimates of $\|\gamma_{N,t} - p_{N,t}\|_{\mathcal{L}^1}$ were given in terms of the number N of electrons and the time t, in the semi-classical mean-field scaling. Their method is based on the Grönwall lemma, similarly to [57,52] in the bosonic case. The second article deals with the semi-relativistic case. The authors pointed out that with a bounded potential, in this scaling, the exchange term in the time-dependent Hartree–Fock equation does not play a role so that the time-dependent Hartree–Fock equation reduces to the time-dependent Hartree equation. In [50], the fermionic Hartree equation in the large volume case is considered by generalizing the method of [52]. Interactions of the form $|x|^{-s}$ are considered, with the corresponding $\lambda = N^{-1+s/3}$. Under the condition that the Hartree–Fock kinetic energy per particle is bounded uniformly in time, a derivation of the TDHF equation is given for $0 < s < 3/5$, and for Coulomb interaction with either a mild singularity cutoff on a ball with radius $N^{-1/6+\varepsilon}$, for any $\varepsilon > 0$, or for the full Coulomb interaction under certain Sobolev conditions on the solution to the TDHF equation which are not proven in this work. Explicit bounds in terms of N, the Hartree–Fock kinetic energy and t are given. Furthermore, in [50], the main result of [16] is reproduced with a different method than in [16] and written down for weaker conditions on the closeness of the initial state to a Slater determinant.

Main estimate of this article (see Theorem 2.1). Given a normalized initial state $\Psi_0 \in \mathfrak{H}_f^{(N)}$ and the one-particle density matrix $p_0 \equiv \gamma_{\Phi_{HF,0}}$ associated with a Slater determinant $\Phi_{HF,0} = \varphi_{1,0} \wedge \cdots \wedge \varphi_{N,0}$, with $\langle \varphi_{i,0}, \varphi_{j,0} \rangle$ being orthonormal orbitals in $\mathrm{H}^1(\mathbb{R}^3)$, γ_t the one-particle density matrix of the solution Ψ_t to (1) and p_t the solution to (11) obey the trace norm estimate

$$\frac{1}{N}\|\gamma_t - p_t\|_{\mathcal{L}^1} \leq \sqrt{8}\sqrt{N^{2/3}\frac{1}{N}\|\gamma_0 - p_0\|_{\mathcal{L}^1}\exp(C_{\lambda,N,K}t) + N^{-1/3}\big(\exp(C_{\lambda,N,K}t) - 1\big)}\,, \tag{14}$$

with $C_{\lambda,N,K} = 30\lambda\sqrt{K}N^{1/6}$, where K is a bound on the kinetic energy of p_t which is assumed to be uniform in time (see (15)).

Discussion of the results. Roughly speaking, the estimate (14) implies that, starting from a state close to a Slater determinant for the N-body Schrödinger equation and from the corresponding one-particle density matrix for the TDHF equation, the Hartree–Fock approximation is justified up to times of order $(\lambda\sqrt{K}N^{1/6})^{-1}$, where K is the kinetic energy (which, for repulsive systems, is bounded by the total energy of the system, uniformly in time) and λ the coupling constant. Hence, our assumption on the initial state is given in terms of energy, and not in the form of "increasing" sequences of Slater determinants. This assumption seems more natural to the authors as it is closer to a thermodynamic assumption on the system. In our proof we obtain a rate of convergence of $N^{-1/6}$. For the initial data, in order to have convergence, we can allow states with $N^{-1/3}\|\gamma_0 - p_0\|_{\mathcal{L}^1} \to 0$ for $N \to \infty$. This means, e.g., that, for any $\varepsilon > 0$, the initial state can have $N^{1/3-\varepsilon}$ particles outside the condensate, i.e., the Slater determinant structure.

The fact that the estimate (14) is relevant when $\lambda N^{1/6}K^{1/2}t$ is of order one, restricts its applicability to a regime where the kinetic energy dominates the direct and exchange terms. This implies that the evolution is the free evolution to leading order. Estimate (14) captures the subleading effect of the direct term on the dynamics and is thus relevant provided that $K \gg N^{4/3}$. We substantiate this by heuristic arguments in Appendix B. Let us stress that estimate (14) requires no additional assumption on the initial states other than the Hartree–Fock kinetic energy to be finite. Furthermore, estimate (14) applies to the repulsive or attractive Coulomb interaction, which is very relevant for many physical systems.

Compared to [32], where also the Coulomb potential was considered, our result holds for larger time scales. In [32], the $\lambda = N^{-1}$-scaling was assumed and, by a rescaling in time and in space, the result also applies to a large neutral atom (i.e., with charge $N \gg 1$ and $\lambda = \alpha$). With the result of [32] the Hartree–Fock approximation is then justified up to times of order N^{-2}. Assuming we have a state with a negative energy, the kinetic energy is controlled by a universal multiple of $N^{7/3}$ (see Section 2 for more details), and our estimate allows us to justify the approximation up to much larger times, of order $N^{-4/3}$. (Note, however, that our estimate deteriorates if the energy of the state is higher.)

Compared to [16] where the semi-classical scaling (8) is considered, our result allows us to control the approximation only up to times of order $N^{-1/3}$, whereas the estimates in [16] allow one to control the approximation up to times of order 1 (however, only for bounded two-body potentials). This comes from the fact that we do not assume any semi-classical structure on the initial data. Note that our strategy is similar to the one of [16] since we do not use the BBGKY hierarchy but instead make use of a Grönwall lemma. An important difference lies in the decomposition of the potential: in [16] a Fourier decomposition is used whereas we use the Fefferman–de la Llave formula.

Let us compare our results to [50] where the mean-field scaling for large volume (7) is considered. Note that there the Schrödinger dynamics is compared to the fermionic Hartree equation without exchange term. While in [50] other interactions are also considered, for Coulomb interaction, essentially two results are proven. First, for regularized Coulomb interaction with singularity cut off on a ball with radius $N^{-1/6+\varepsilon}$ for any $\varepsilon > 0$, convergence of the Schrödinger dynamics to the fermionic Hartree dynamics is shown in terms of a counting measure α_g, with convergence rate depending on the cutoff. Note, that we use the same measure in our proof, see also Remark 3.5, but we formulate our main result only in terms of the trace norm difference of reduced densities. The improvement of our result is that it holds for full Coulomb interaction without any regularization and, in general, with a better convergence rate. For the second result in [50] a bound on $\mathrm{Tr}[(-\Delta)^{3+\varepsilon}p_t]$ is assumed. Under that condition convergence for full Coulomb interaction in terms of α_g and the trace norm difference is shown, with rate $N^{-1/2}$ in the trace norm sense. This bound on $\mathrm{Tr}[(-\Delta)^{3+\varepsilon}p_t]$ was, however, not proven to hold for $t > 0$. Compared to that, our result holds for any initial

condition with kinetic energy bounded by CN, without further assumptions, but only with a convergence rate of $N^{-1/6}$ in the trace norm sense.

Sketch of our derivation of estimates on the accuracy of the TDHF approximation. We derive an estimate on the trace norm of the difference $\gamma_t - p_t$ between the one-particle density matrix $\gamma_t \equiv \gamma_{\Psi_t}$ of the (full) solution $\Psi_t = e^{-itH}\Psi_0$ of (1) and the one-particle density matrix p_t solving the TDHF equation (11). Our work is inspired by Pickl [52], where one of us developed a new method for bosonic systems which was generalized to fermion systems in [50] by two of us. The method uses a Grönwall estimate for a well-chosen quantity called the *number of bad particles* in [52]. We refer to the quantity we chose to control as the *degree of evaporation* S_g. The subscript g refers to a freedom in the choice of a weight function g which allows us to fine-tune the distance of γ_t (the density matrix of Ψ_t) to p_t in a suitable way. For the simplest choice $g(x) = x$, S_g is called the *degree of non-condensation* in [38, Remark (a) on p. 5], while in [59] it is called *Verdampfungsgrad*, which translates to *degree of evaporation*.

We show that the degree of evaporation S_g is directly related to the trace norm $\|\gamma - p\|_{\mathcal{L}^1}$. We then calculate the time derivative of S_g and split it into three terms that we estimate separately. To obtain the estimates we make use of correlation inequalities which may be seen to be a dynamical version of the correlation estimate presented in [7]. (See also [38] for an alternative proof of that correlation estimate which does not make use of second quantization.) While we estimate two of the terms in a way very similar to [50], our estimate for the remaining term (here called $\mathcal{A}$; in [50] called (I)) is very different and allows us to treat the full Coulomb potential. This term is physically the most important, since its smallness is a consequence of cancellations between the Hartree–Fock and the many-body interaction. The bounds on this term are the key estimates of this work. They are obtained by using the Fefferman–de la Llave decomposition formula [31]. We remark that, in view of the generalization of this decomposition derived in [39,37], our result applies to a more general class of two-body interaction potentials. The Lieb–Thirring inequality [46] and Hardy's inequality then provide an estimate in terms of kinetic energy. Finally, we note that in many physically relevant cases the estimate in terms of kinetic energy can be stated in terms of an estimate on the initial total energy of the system.

Outline of the article. In Section 2 we state our main result, along with applications to molecules or the mean-field limit. In Section 3 we introduce the degree of evaporation S_g and relate it to the difference between the one-particle density matrix of the solution to our model and the solution to the TDHF equation. We then calculate the time derivative of S_g and provide bounds for the different contributions, thus proving our main theorem. In Appendix A we recall some results about the theory of the TDHF equation.

2. Main result and applications

Our main result is an estimate of the trace norm $\|\cdot\|_{\mathcal{L}^1}$ of the difference between the one-particle density matrix of the solution to the many-body Schrödinger equation (1) and the solution to the time-dependent Hartree–Fock equation (11) in terms of the kinetic energy of the system. As usual, we denote by $\mathrm{H}^1(\mathbb{R}^3)$ the Sobolev space of weakly differentiable functions with square-integrable derivative.

We henceforth make use of the following notation:

- Let $\Psi_0 \in \mathfrak{H}_f^{(N)}$ be a normalized initial state, and let $\gamma_t := \gamma_{\Psi_t}$ be the one-particle density matrix of the solution $\Psi_t = e^{-iHt}\Psi_0$ to the Schrödinger equation (1) with Hamiltonian H from (2) (i.e., with Coulomb interaction).
- Let $\Phi_{HF,0} = \varphi_{1,0} \wedge \cdots \wedge \varphi_{N,0}$ be a Slater determinant, with $\varphi_{j,0} \in \mathrm{H}^1(\mathbb{R}^3)$ and $\langle \varphi_{j,0}, \varphi_{k,0} \rangle_{\mathfrak{h}} = \delta_{jk}$, for $1 \leq j, k \leq N$. Let $p_0 := \gamma_{\Phi_{HF,0}}$ be the one-particle density matrix of $\Phi_{HF,0}$ and p_t be the solution to the time-dependent Hartree–Fock equation (11) with initial condition p_0.

Theorem 2.1. *Assume that the kinetic energy of p_t is uniformly bounded in time,*

$$K \;:=\; \sup_{t\geq 0} \mathrm{Tr}[-\Delta p_t] \;<\; \infty\,. \tag{15}$$

Under the assumption of (15) the estimate

$$\frac{1}{N}\|\gamma_t - p_t\|_{\mathcal{L}^1} \leq \sqrt{8}\sqrt{N^{2/3}\frac{1}{N}\|\gamma_0 - p_0\|_{\mathcal{L}^1}\exp(C_{\lambda,N,K}t) + N^{-1/3}\big(\exp(C_{\lambda,N,K}t) - 1\big)} \tag{16}$$

holds true with $C_{\lambda,N,K} = 30\lambda\sqrt{K}N^{1/6}$.

The proof of Theorem 2.1 is postponed to Section 3.

Remark 2.2. One of the ingredients of our proof is the Fefferman–de la Llave decomposition of the Coulomb potential [31]

$$\frac{1}{|x|} \;=\; \int_0^\infty \frac{16}{\pi\, r^5}\,(1_{B(0,r/2)} * 1_{B(0,r/2)})(x)\,dr\,, \tag{17}$$

an identity that holds for all $x \in \mathbb{R}^3 \setminus \{0\}$, where $1_{B(0,r/2)}$ is the characteristic function of the ball of radius $r/2$ centered at the origin in $\mathbb{R}^3$. A generalization of this decomposition to a class of two-body interaction potentials v of the form

$$v(x) \;=\; \int_0^\infty g_v(r)\,(1_{B(0,r/2)} * 1_{B(0,r/2)})(x)\,dr\,, \tag{18}$$

with $x \in \mathbb{R}^3 \setminus \{0\}$, was given in [39] under Assumption 2.3 below, and our proof largely generalizes to those potentials v. More precisely, the assertion of Theorem 2.1 holds true and without any change in the constants, if we replace the Coulomb potential by any pair potential v that satisfies Assumption 2.4 below, which in particular implies $v(x) \leq |x|^{-1}$. Note that the assumption of semi-boundedness of v is only used to ensure the global existence of a solution to the TDHF equation. One could drop it to study problems up to the time the solution to the TDHF blows up.

Assumption 2.3. *The function $v : \mathbb{R}^3 \setminus \{0\} \to \mathbb{R}$ has the following properties:*

- *v is a radial function, and there exists a function $\tilde{v} \in C^3[(0,\infty);\mathbb{R}]$ such that $v(x) = \tilde{v}\big(|x|\big)$, for all $x \in \mathbb{R}^3 \setminus \{0\}$,*
- *$r^m \frac{d^m\tilde{v}}{dr^m}(r) \to 0$, as $r \to \infty$, for $m = 0, 1, 2$,*
- *$\lim_{R\to\infty}\int_1^R r^3\, g_v(r)\,dr$ exists, with $g_v(r) := \frac{2}{\pi}\frac{d}{dr}\big(\frac{1}{r}\frac{d^2\tilde{v}}{dr^2}(r)\big)$.*

Note that $g_{|\cdot|^{-1}}(r) = \frac{16}{\pi}r^{-5}$ in case of the Coulomb potential which is prototypical for the following further assumption:

Assumption 2.4. *(With the same notation as in Assumption 2.3.) The function $v : \mathbb{R}^3 \to \mathbb{R}$ satisfies Assumption 2.3, $|g_v(r)| \leq \frac{16}{\pi}r^{-5}$ and, for some $\mu \in \mathbb{R}$, $v(x) \geq \mu$ for all x.*

Remark 2.5. Note that we actually prove a slightly stronger result in Theorem 3.8 in terms of the degree of evaporation $S_g(t)$ (with properly chosen g), which is defined in Definition 3.3. The way our result is formulated in Theorem 3.8 can directly be compared to the results in [50].

Remark 2.6. Note that the two summands in the square root on the right-hand side of (16) come from different contributions which we call $\mathcal{A}_t$, $\mathcal{B}_t$ and $\mathcal{C}_t$ (and which are called (I), (II), (III) in [50]), see Proposition 3.9. It is interesting to note that all three terms contribute to the first summand (which is proportional to $\|\gamma_0 - p_0\|_{\mathcal{L}^1}$) but only the $\mathcal{B}_t$ term contributes to the second summand.

Remark 2.7. Note that it is sufficient to prove Theorem 2.1 with Ψ_0 in $\mathfrak{H}_f^{(N)} \cap \mathrm{H}^1(\mathbb{R}^3)^{\otimes N}$. A density argument then provides the result for a general Ψ_0 in $\mathfrak{H}_f^{(N)}$.

Let us discuss some cases when the assumption that the Hartree–Fock kinetic energy is uniformly bounded in time is satisfied. In Propositions 2.11 and 2.12 we give explicit bounds on the kinetic energy K in terms of the energy expectation value $\langle \Phi_{HF,0}, H\Phi_{HF,0}\rangle$ of the initial state $\Phi_{HF,0}$ and the ground state energy for examples presented in Section 1. In the case of atoms or molecules this follows from known estimates, which we now recall.

To formulate these, we denote the energy expectation value and the kinetic energy expectation value of a normalized wave function $\Psi \in \mathfrak{H}_f^{(N)} \cap \mathrm{H}^1(\mathbb{R}^3)^{\otimes N}$ by

$$\mathcal{E}(\Psi) = \langle \Psi, H\Psi\rangle \quad \text{and} \quad \mathcal{K}(\Psi) = \left\langle \Psi, \Big(\sum_{j=1}^{N} -\Delta_j\Big)\Psi \right\rangle .$$

For atoms and molecules the ground state energy E_{gs} is defined as

$$E_{gs} = \inf \Big\{ \mathcal{E}(\Psi) \;\Big|\; \Psi \in \mathfrak{H}_f^{(N)} \cap \mathrm{H}^1(\mathbb{R}^3)^{\otimes N},\ \|\Psi\|_{\mathfrak{H}_f^{(N)}} = 1\ , \\ R_1, \dots, R_M \in \mathbb{R}^3, l \neq m \Rightarrow R_l \neq R_m \Big\} .$$

Equipped with this notation, we formulate the coercivity of the energy functional on the Sobolev space of states with finite kinetic energy:

Proposition 2.8. *Consider a neutral atom or a molecule as in (3) or (4). If $E_{gs} \leq 0$ then*

$$\mathcal{K}(\Psi) \leq \Big(\sqrt{\mathcal{E}(\Psi) - E_{gs}} + \sqrt{-E_{gs}}\Big)^2 \leq 2\mathcal{E}(\Psi) + 4|E_{gs}| .$$

Proof. See [45, p. 132]. □

Using Proposition 2.8 along with the conservation of the total energy for both the Schrödinger equation and the TDHF equation (see Appendix A) we get the following bound on the kinetic energy.

Proposition 2.9. *Assume that $\Phi_{HF,0} = \varphi_{1,0} \wedge \cdots \wedge \varphi_{N,0}$ is a Slater determinant, with $\varphi_{j,0} \in \mathrm{H}^1(\mathbb{R}^3)$ and $\langle \varphi_{j,0}, \varphi_{k,0}\rangle_{\mathfrak{h}} = \delta_{jk}$, for $1 \leq j,k \leq N$. Then, in the case of atoms or molecules as in (3) or (4),*

$$K \;:=\; \sup_{t\geq 0} \mathrm{Tr}[-\Delta p_t] \;\leq\; \Big(\sqrt{\mathcal{E}(\Phi_{HF,0}) - E_{gs}} + \sqrt{-E_{gs}}\Big)^2 . \tag{19}$$

Thus, if $\mathcal{E}(\Phi_{HF,0}) \leq 0$ then

$$K \;\leq\; -4E_{gs} . \tag{20}$$

We also recall a known bound for the ground state energy, see [46] or [45], whose units we use.

Proposition 2.10 *(Ground state energy of a molecule). For a molecule with nuclei of charges $Z_1, \dots, Z_M > 0$ at pairwise distinct positions $R_1, \dots, R_M \in \mathbb{R}^3$, with $\lambda = \alpha$, $\nu = \sum_{m<l} \alpha Z_m Z_l / |R_m - R_l|$ as in (4), and $Z = \max\{Z_1, \dots, Z_M\}$, the ground state energy satisfies the bound*

$$0 \;<\; -E_{gs} \;\leq\; (0.231)\alpha^2 N \left[1 + 2.16\, Z \Big(\frac{M}{N}\Big)^{1/3}\right]^2 .$$

Proposition 2.11 *(Neutral atom). In case of an atom with $N = Z$ the ground state energy satisfies*

$$0 \;<\; -E_{gs} \;\leq (2.31)\alpha^2\, N^{7/3} .$$

Proposition 2.12 *(TDHF equations without external potential and with repulsive interaction). For $h^{(1)} = -\Delta/2$ and $v(x) = |x|^{-1}$, the Hartree–Fock kinetic energy is bounded by the total Hartree–Fock energy (for any $\lambda > 0$), which is preserved in time, i.e.,*

$$K \;\leq\; \mathcal{E}(\Phi_{HF,0}) .$$

Finally, let us note that for attractive Coulomb interaction without external field, we have the bound

$$K \leq 2\mathcal{E}(\Phi_{HF,0}) + C\lambda^2 N^{7/3} , \tag{21}$$

which follows from the Lieb–Thirring inequality and which we prove in Appendix B. Thus, also for attractive interaction, the bounds $K \leq CN$ in the mean-field scaling for large volume (7) and $K \leq CN^{5/3}$ in the semi-classical mean-field scaling (8) hold, if the corresponding bounds hold for the total energy.

3. Control of the degree of evaporation S_g

We first introduce the degree of evaporation S_g, which is a function of a state on the Fock space and a one-particle density matrix. We use S_g as an indicator of closeness of the Hartree–Fock to the Schrödinger quantum state.

3.1. Definition and properties of the degree of evaporation

For A and more generally $B^{(M)}$ $(M \leq N)$ linear operators acting on $\mathfrak{h}$ and $\mathfrak{H}_f^{(M)}$, respectively, we use the notation

$$\mathrm{d}\Gamma(A) := \sum_{j=1}^{N} A_j \qquad \text{and} \qquad \mathrm{d}\Gamma^{(M)}(B^{(M)}) := \sum_{\substack{j_1,\dots,j_M=1 \\ j_1 \neq j_2 \cdots \neq j_M}}^{N} B^{(M)}_{j_1 \cdots j_M} , \tag{22}$$

as operators on $\mathfrak{H}_f^{(N)}$, with A_j acting on the j^{th} factor in $\mathfrak{h}^{\otimes N}$ and $B^{(M)}$ acting on the $j_1^{th}, \dots, j_M^{th}$ factors in $\mathfrak{h}^{\otimes N}$, respectively.

Remark 3.1. Although we do not use the fermion creation and annihilation operators a^*, a, note that (22) coincides with the second quantization $\mathrm{d}\Gamma$ in quantum field theory in the sense that

$$\mathrm{d}\Gamma(A) = \int A(x;y)\; a^*(x) a(y)\; dxdy ,$$

or, more exactly, its restriction to the N-particle sector. Similarly, e.g.,

$$\mathrm{d}\Gamma^{(2)}(B^{(2)}) = \int B^{(2)}(x_1, x_2; y_1, y_2)\; a^*(x_2)a^*(x_1)a(y_1)a(y_2)\; dx_1 dx_2 dy_1 dy_2 \;.$$

Let $\mathcal{L}^1(\mathfrak{H})$ denote the space of trace class operators on a Hilbert space $\mathfrak{H}$. We use the partial trace $\mathrm{Tr}_2 : \mathcal{L}^1(\mathfrak{h}^{\otimes 2}) \to \mathcal{L}^1(\mathfrak{h})$ which is defined for $B^{(2)} \in \mathcal{L}^1(\mathfrak{h}^{\otimes 2})$ to be the operator $\mathrm{Tr}_2[B^{(2)}] \in \mathcal{L}^1(\mathfrak{h})$ such that

$$\mathrm{Tr}\left[\,\mathrm{Tr}_2(B^{(2)})\,A\right] \;=\; \mathrm{Tr}\left[B^{(2)}\,(A\otimes \mathbb{1}_{\mathfrak{h}})\right] \tag{23}$$

holds for all $A \in \mathcal{B}(\mathfrak{h})$.

Definition 3.2. For an N-particle density matrix $\rho \in \mathcal{L}^1_+(\mathfrak{H}_f^{(N)})$, i.e., a non-negative trace-class operator on $\mathfrak{H}_f^{(N)}$ of unit trace, the one- (resp. two-)particle density matrix of ρ is denoted by γ_ρ (resp. $\gamma_\rho^{(2)}$). It is the operator on $\mathfrak{h}$ (resp. $\mathfrak{H}_f^{(2)}$) such that

$$\forall A \in \mathcal{B}(\mathfrak{h}): \quad \mathrm{Tr}_{\mathfrak{H}_f^{(N)}}[\rho\,\mathrm{d}\Gamma(A)] \;=\; \mathrm{Tr}_{\mathfrak{h}}[\gamma_\rho\,A]\,, \tag{24}$$

$$\forall B^{(2)} \in \mathcal{B}(\mathfrak{H}_f^{(2)}): \quad \mathrm{Tr}_{\mathfrak{H}_f^{(N)}}\left[\rho\,\mathrm{d}\Gamma^{(2)}(B^{(2)})\right] \;=\; \mathrm{Tr}_{\mathfrak{H}_f^{(2)}}\left[\gamma_\rho^{(2)}\,B^{(2)}\right]. \tag{25}$$

We note that γ_ρ and $\gamma_\rho^{(2)}$ satisfy

$$0 \le \gamma_\rho \le \mathbb{1}\,,\quad \mathrm{Tr}_{\mathfrak{h}}[\gamma_\rho] = N\,,\quad 0 \le \gamma_\rho^{(2)} \le N\,,\quad \mathrm{Tr}_{\mathfrak{H}_f^{(2)}}[\gamma_\rho^{(2)}] = N(N-1) \tag{26}$$

(see, e.g., [9, Theorem 5.2]). Further note that we are slightly abusing notation since the one-particle density matrix was defined for wave functions in Eq. (13), rather than for density matrices. We thus identify $\gamma_\Psi \equiv \gamma_{|\Psi\rangle\langle\Psi|}$, for all normalized $\Psi \in \mathfrak{H}_f^{(N)}$, whenever this does not lead to confusion.

Definition 3.3. Let $N \in \mathbb{N}$ and

$$\mathfrak{S}_N \;:=\; \left\{\eta \in \mathcal{L}^1(\mathfrak{h}) \,\middle|\, 0 \le \eta \le \mathbb{1}\,,\ \mathrm{Tr}[\eta] = N\right\},$$

and g be a continuous function from $\mathbb{R}$ to $\mathbb{R}$. The map $S_g : \mathcal{L}^1_+(\mathfrak{H}_f^{(N)}) \times \mathfrak{S}_N \to \mathbb{R}_0^+$ defined by

$$S_g(\rho,\eta) \;:=\; \mathrm{Tr}[\rho\;\hat{g}]\,, \tag{27}$$

where $\hat{g} := g\big(\mathrm{d}\Gamma(\mathbb{1}-\eta)\big)$, is called the degree of evaporation (of ρ relative to η). The translation of g by $k \in \mathbb{Z}$, is denoted by $\tau_k g(y) := g(y-k)$.

Note that when the expression of g is too long to fit under a hat, we write $(g)^\wedge$ instead of $\hat{g}$.

Remark 3.4. $\hat{g} := g\big(\mathrm{d}\Gamma(\mathbb{1}-\eta)\big)$ is defined using the functional calculus [54,56].

Remark 3.5. The particular case in which η is a rank-N projector is of importance in the sequel, and we then write

$$p := \eta \qquad \text{and} \qquad q := \mathbb{1} - \eta\,.$$

In this case, only the values of g on $\{0,\dots,N\}$ are relevant for the definition of $\hat{g}$ and the continuity assumption on g can be dropped. We can then give an alternative and equivalent viewpoint using the orthogonal projections

$$P_m^{(M)} := \sum_{\substack{a\in\{0,1\}_0^M \\ |a|=m}} \bigotimes_{\ell=1}^{M} \big((1-a_\ell)p + a_\ell q\big) = 1_{\{m\}}\big(\mathrm{d}\Gamma(q)\big)$$

on $\mathfrak{h}^{\otimes M}$, with $|a| = a_1 + \cdots + a_M$, for $M \in \mathbb{N}$ and $m \in \mathbb{Z}$. (Note that with this definition $P_m^{(M)} = 0$ for $m \notin \{0, \ldots, M\}$.) We can then write down the spectral decomposition of $\mathrm{d}\Gamma(q)$,

$$\mathrm{d}\Gamma(q) = \sum_{n\in\mathbb{Z}} n\, P_n^{(N)}\,, \tag{28}$$

i.e., $P_n^{(N)}$ is the projection on the eigenspace of $\mathrm{d}\Gamma(q)$ associated with the eigenvalue $n \in \mathbb{Z}$. It follows that

$$S_g(\rho, p) = \sum_{n=0}^{N} g(n)\ \mathrm{Tr}\left[\rho P_n^{(N)}\right]. \tag{29}$$

In this form we see directly that for $\rho = |\Psi\rangle\langle\Psi|$ we have $S_g = \alpha_g$, where α_g is the functional used in [50, Definition 2.1] to control the closeness of a Hartree–Fock state to a Schrödinger state. However, note that there is a difference in the choice of normalization. The particular choices of functions, f in [50] and g in our article, are related through $g = N f$, such that $S_g = N\alpha_f$. Note also that for $g(x) = x$ we find $S_{Id_{\mathbb{R}}}(\rho, p) = \mathrm{Tr}[\gamma_\rho(\mathbb{1} - p)]$.

Remark 3.6. For $g(x) = x$, the functional S_g has been used in [8,38,59] in the context of mean-field approximations for ground states. In [38], S_g is called "degree of non-condensation" or "the relative number of particles outside the Fermi sea". For general g, a bosonic variant of S_g was introduced for the derivation of mean-field dynamics in [52] and for the derivation of the NLS equation in [51,53]. For the derivation of mean-field dynamics for fermions, S_g was introduced in [50]. Note that for $g(x) = x$ and $\Psi_0 \in \mathfrak{H}_f^{(N)}$, $2S_g(t)$ coincides with the quantity $\langle \mathcal{U}_N(t;0)\xi, \mathcal{N}\mathcal{U}_N(t;0)\xi\rangle$ in [16] in case $\xi = R^*_{\nu_N}\Psi_0$.

Let us collect some properties of $S_g(\rho, \eta)$ and show how it relates to the distance of γ_ρ to η in trace norm. (Note that some of the statements were already proven in [16] and [50].) We denote the Hilbert–Schmidt norm by $\|\cdot\|_{\mathcal{L}^2}$.

Proposition 3.7. *For $\eta \in \mathfrak{S}_N$ and ρ a density matrix with one particle density matrix γ, the degree $S_g(\rho, \eta)$ of evaporation has the properties*

$$\inf_{0\le x\le N} g(x) \le S_g\ (\rho, \eta) \le \sup_{0\le x\le N} g(x)\,, \tag{30}$$

$$\|\gamma - \eta\|^2_{\mathcal{L}^2}\ \le 2S_{Id_{\mathbb{R}}}(\rho, \eta)\,, \tag{31}$$

$$g_1 \le g_2\ \textit{on}\ [0, N] \Rightarrow S_{g_1}(\rho, \eta) \le S_{g_2}(\rho, \eta)\,, \tag{32}$$

$$g \mapsto S_g(\rho, \eta)\ \ \textit{is linear,} \tag{33}$$

for g, g_1, g_2 functions from $\mathbb{R}$ to $\mathbb{R}$.

If furthermore $p^2 = p$ is a rank-N orthogonal projection and $g(0) = 0$, $g(x) \ge x$ on $[0, N]$, then

$$\frac{1}{N}\|\gamma - p\|_{\mathcal{L}^1}\ \le\ \sqrt{\frac{8}{N}\, S_g(\rho, p)}\,, \tag{34}$$

$$S_g(\rho, p) \le \sup_{0<x\le N} \left|\frac{g(x)}{x}\right| \|\gamma - p\|_{\mathcal{L}^1}\,. \tag{35}$$

Proof. The spectrum of $\mathrm{d}\Gamma(q)$ (restricted to $\mathfrak{H}_f^{(N)}$) is included in $[0, N]$, thus

$$\inf_{0\leq x\leq N} g(x) \leq \hat{g} \leq \sup_{0\leq x\leq N} g(x)\,,$$

in the sense of quadratic forms. As ρ is a state, Eq. (30) follows.

Equation (31) follows from

$$\begin{aligned}\|\gamma-\eta\|_{\mathcal{L}^2}^2 &= \mathrm{Tr}\left[(\gamma-\eta)^2\right] = \mathrm{Tr}\left[\gamma^2+\eta^2-2\gamma\eta\right]\\ &= 2S_{Id_{\mathbb{R}}}(\rho,\eta) - \mathrm{Tr}[\gamma-\gamma^2] - \mathrm{Tr}[\eta-\eta^2] \leq 2S_{Id_{\mathbb{R}}}(\rho,\eta)\,.\end{aligned}$$

Equations (32) and (33) follow from the properties of the functional calculus.

For the proof of (34), we first remark that $\gamma - p$ has at most N negative eigenvalues (counting multiplicities). This is a well-known consequence of $\gamma - p \geq -p$ and the fact that p is a rank-N orthogonal projection (see, e.g., [55]), but we include its proof for the sake of completeness: Suppose that $\gamma - p$ has at least $N+1$ negative eigenvalues. Then there is a subspace W of dimension $N+1$ such that $\langle\varphi|(\gamma-p)\varphi\rangle < 0$, for all $\varphi \in W \setminus \{0\}$. Since $\gamma \geq 0$, this implies that $\langle\varphi|p\varphi\rangle > 0$, for all $\varphi \in W \setminus \{0\}$. On the other hand, the largest dimension of a subspace with this property is N, by the minmax principle and the fact that p has precisely N positive eigenvalues, which contradicts the existence of W.

Denoting the number of negative eigenvalues (counting multiplicities) of $\gamma - p$ by M, we consequently have that $M \leq N$. Let $\lambda_1, \ldots, \lambda_M$ be these M negative eigenvalues of $\gamma - p$, and $\lambda_{M+1}, \lambda_{M+2}, \ldots$ be the non-negative ones. Since $\mathrm{Tr}[\gamma - p] = 0$, it follows that

$$-(\lambda_1 + \cdots + \lambda_M) = \sum_{m=M+1}^{\infty} \lambda_m\,.$$

Using the Cauchy–Schwarz inequality and $M \leq N$, we obtain

$$\begin{aligned}\|\gamma-p\|_{\mathcal{L}^1} &= \sum_{m=M+1}^{\infty}\lambda_m - \sum_{m=1}^{M}\lambda_m = -2\sum_{m=1}^{M}\lambda_m \leq 2\sqrt{M}\Big(\sum_{m=1}^{M}\lambda_m^2\Big)^{1/2}\\ &\leq 2\sqrt{N}\Big(\sum_{m=1}^{\infty}\lambda_m^2\Big)^{1/2} = 2\sqrt{N}\|\gamma-p\|_{\mathcal{L}^2}\,,\end{aligned}$$

and Eq. (34) follows from Eq. (31) and $S_{Id_{\mathbb{R}}}(\rho,p) \leq S_g(\rho,p)$.

To prove Eq. (35), we observe that $g(x) \leq \sup_{0<x\leq N}\left\{\left|\frac{g(x)}{x}\right|\right\}x$ on $[0, N]$ and thus, using positivity preservation and linearity, we have

$$S_g(\rho,p) \leq \sup_{0<x\leq N}\left\{\left|\frac{g(x)}{x}\right|\right\} S_{Id_{\mathbb{R}}}(\rho,p)\,.$$

We conclude with

$$S_{Id_{\mathbb{R}}}(\rho,p) = \mathrm{Tr}[\gamma(1-p)] = \mathrm{Tr}[p(p-\gamma)p] \leq \|\gamma-p\|_{\mathcal{L}^1}\,,$$

using again $p = p^2$ and $\mathrm{Tr}[\gamma] = N = \mathrm{Tr}[p]$. □

Let us now state the main result of this section. Recall that we defined $K := \sup_{t\geq 0}\mathrm{Tr}[-\Delta p_t]$.

Theorem 3.8. *Assume (15) holds, i.e., the kinetic energy is uniformly bounded, as in Theorem 2.1. Then, writing* $\rho_t = |\Psi_t\rangle\langle\Psi_t|$,

$$S_{g_{1/3}}(\rho_t, p_t) \leq S_{g_{1/3}}(\rho_0, p_0) \exp\left(30\lambda\sqrt{K}N^{1/6}t\right) + N^{2/3}\Big(\exp\left(30\lambda\sqrt{K}N^{1/6}t\right) - 1\Big)\,, \tag{36}$$

where, for $\theta > 0$, g_θ *is the function from* $\mathbb{R}$ *to* $\mathbb{R}$ *defined by*

$$\forall x \in \mathbb{R}\,, \quad g_\theta(x) := N^{1-\theta} x 1_{[0,N^\theta]}(x) + N 1_{(N^\theta,\infty)}(x)\,. \tag{37}$$

Note that the function g_θ was also used to obtain the results in [50]. Theorem 3.8 will be proved in the following subsections. The strategy is to obtain a bound for dS_t/dt in terms of S_t and N^δ, for some $\delta < 1$, and then integrate it, in the spirit of the Grönwall lemma.

Before we turn to the proof of Theorem 3.8, we show how Theorem 3.8 and the properties of the degree of evaporation imply Theorem 2.1, the main result of this article.

Proof of Theorem 2.1. Since $g_{1/3} \geq Id_{\mathbb{R}}$ on $[0, N]$, we can apply Eq. (34) to Eq. (36) which gives

$$\frac{1}{N}\|\gamma_t - p_t\|_{\mathcal{L}^1} \leq \sqrt{\frac{8}{N}}\sqrt{S_{g_{1/3}}(\rho_0, p_0)\exp(30\lambda\sqrt{K}N^{1/6}t) + N^{2/3}\big(\exp(30\lambda\sqrt{K}N^{1/6}t) - 1\big)}\,.$$

Equation (35) with $g_{1/3}$ yields $S_{g_{1/3}}(\rho, p) \leq N^{2/3}\|\gamma - p\|_{\mathcal{L}^1}$ which then gives Eq. (16). □

The rest of this section is devoted to the proof of Theorem 3.8.

3.2. Time-derivative of the degree of evaporation

In this subsection we calculate the time derivative of the degree of evaporation $S_g(t) := S_g(\rho_t, p_t)$ and bring it into a form that can be conveniently estimated. Then most of the following subsections provide bounds on the different contributions to the time derivative. First, recall the Fefferman–de la Llave decomposition, for $x \neq y \in \mathbb{R}^3$,

$$\frac{1}{|x-y|} = \int_{\mathbb{R}^3} d^3z \int_0^\infty \frac{dr}{\pi\, r^5}\, X_{r,z}(x)\, X_{r,z}(y)\,, \tag{38}$$

of the Coulomb potential, where $X_{r,z}(x) := 1_{|x-z|\leq r}$ is the characteristic function of the ball in $\mathbb{R}^3$ of radius $r > 0$ centered at $z \in \mathbb{R}^3$. This formula can also be written as

$$v^{(2)} = \int d\mu(\omega)\, X_\omega \otimes X_\omega\,, \tag{39}$$

where $\omega = (r, z) \in \mathbb{R}^+ \times \mathbb{R}^3$ and $\int d\mu(\omega)\, f(\omega) := \int_{\mathbb{R}^3} d^3z \int_0^\infty \frac{dr}{\pi\, r^5}\, f(r, z)$. The form (38) is convenient for the estimates derived below, but we note that it agrees with (18), of course. Note that the terms $\mathcal{A}_t$, $\mathcal{B}_t$, $\mathcal{C}_t$ in the following proposition are the same as (I), (II), (III) in [50, Lemma 6.5]. However, an important difference lies in the presentation of the $\mathcal{A}_t$ term using the decomposition (38), which enables us to handle the case of the Coulomb interaction. In the following, $\Im$ denotes the imaginary part.

Proposition 3.9. *For all monotonically increasing* $g : \mathbb{R} \to \mathbb{R}$, *the time-derivative of* $S_g(t) = S_g(\rho_t, p_t)$ *(with the notation from Theorems 2.1 and 3.8) is*

$$\frac{dS_g(t)}{dt} = \lambda\big(\mathcal{A}_t + \mathcal{B}_t + \mathcal{C}_t\big)\,, \tag{40}$$

where

$$\mathcal{A}_t := \int 2\Im\,\mathrm{Tr}\left[\mathrm{d}\Gamma(q_t X_\omega p_t)\big(\mathrm{d}\Gamma(p_t X_\omega p_t) - \mathrm{Tr}[X_\omega p_t]\big)\rho_t^{[-1,1]}\right] d\mu(\omega)\,, \tag{41}$$

$$\mathcal{B}_t := \Im\,\mathrm{Tr}\left[\mathrm{d}\Gamma^{(2)}\big((q_t\otimes q_t)v^{(2)}(p_t\otimes p_t)\big)\,\rho_t^{[-2,2]}\right], \tag{42}$$

$$\mathcal{C}_t := 2\Im\,\mathrm{Tr}\left[\mathrm{d}\Gamma^{(2)}\big((q_t\otimes q_t)v^{(2)}(p_t\otimes q_t)\big)\,\rho_t^{[-1,1]}\right], \tag{43}$$

with $q_t := \mathbb{1} - p_t$, *and*

$$\rho^{[-j,j]} := (\tau_{-j}g - g)^{1/2\wedge}\,\rho(g-\tau_j g)^{1/2\wedge}\ . \tag{44}$$

Before we turn to the proof we note that

$$q_t\,p_t \;=\; p_t\,q_t \;=\; 0\,, \tag{45}$$

since p_t is a projection. We further note that, for A, B linear and bounded operators on $\mathfrak{h}$, we have that

$$\mathrm{d}\Gamma(A)\,\mathrm{d}\Gamma(B) \;=\; \mathrm{d}\Gamma^{(2)}(A\otimes B) + \mathrm{d}\Gamma(AB)\,. \tag{46}$$

To prove Proposition 3.9 we need several lemmas. We begin with an evolution equation for $\hat{g}$.

Lemma 3.10. *For any function* $g:\mathbb{R}\to\mathbb{R}$, *with* $\hat{g} = g\big(\mathrm{d}\Gamma(q_t)\big)$ *and* $h_{HF}^{(1)}$ *defined in Eq. (12),*

$$i\partial_t\hat{g} = \big[\mathrm{d}\Gamma\big(h_{HF}^{(1)}(p_t)\big),\hat{g}\big]\,. \tag{47}$$

Proof. First observe that only the values of g on the spectrum of $\mathrm{d}\Gamma(q_t)$ are used to define $\hat{g}$. As the spectrum of $\mathrm{d}\Gamma(q_t)$ is independent of the time t, we could as well consider a time-independent polynomial which coincides with g on the spectrum of $\mathrm{d}\Gamma(q_t)$. It is then enough to prove that Eq. (47) holds for any monomial $\mathrm{d}\Gamma(q_t)^n$. It indeed holds for $n=1$:

$$i\partial_t\mathrm{d}\Gamma(q_t) = \mathrm{d}\Gamma(i\partial_t q_t) = \mathrm{d}\Gamma\big([h_{HF}^{(1)}(p_t),q_t]\big) = \big[\mathrm{d}\Gamma\big(h_{HF}^{(1)}(p_t)\big),\mathrm{d}\Gamma(q_t)\big]\,.$$

Then, for any $n\in\mathbb{N}$,

$$\begin{aligned} i\partial_t\big(\mathrm{d}\Gamma(q_t)\big)^n &= \sum_{j=1}^{n}\big(\mathrm{d}\Gamma(q_t)\big)^{j-1}\ \big[\mathrm{d}\Gamma\big(h_{HF}^{(1)}(p_t)\big),\mathrm{d}\Gamma(q_t)\big]\ \big(\mathrm{d}\Gamma(q_t)\big)^{n-j} \\ &= \big[\mathrm{d}\Gamma\big(h_{HF}^{(1)}(p_t)\big),\big(\mathrm{d}\Gamma(q_t)\big)^n\big]\,, \end{aligned}$$

as all the terms but two simplify in the sum. □

Next, we need a commutation relation (analogous to [50, Lemma 6.4]) involving $\hat{g}$ that enables us to write the time derivative of $S_g(t)$ in terms of a discrete derivative of g. Recall that $\tau_{j-k}g(x) = g(x-j+k)$.

Lemma 3.11. *For integers* $0\le j,k\le M\le N$, *any function* $g:\mathbb{R}\to\mathbb{R}$, *with the notations of Definition 3.3, and* $h^{(M)}\in\mathcal{L}(\mathfrak{h}^{\otimes M})$,

$$\mathrm{d}\Gamma^{(M)}\big(P_j^{(M)}h^{(M)}P_k^{(M)}\big)\,\hat{g} = \widehat{\tau_{j-k}g}\,\mathrm{d}\Gamma^{(M)}\big(P_j^{(M)}h^{(M)}P_k^{(M)}\big)\,. \tag{48}$$

Proof. First, note that if

$$\mathrm{d}\Gamma^{(M)}\big(P_j^{(M)}h^{(M)}P_k^{(M)}\big)\mathrm{d}\Gamma(q) = \widehat{\tau_{j-k}Id}\,\mathrm{d}\Gamma^{(M)}\big(P_j^{(M)}h^{(M)}P_k^{(M)}\big)$$

holds, then Eq. (48) follows by the same argument as in the proof of Lemma 3.10. Using $P_{m_1}^{(M)}P_{m_2}^{(M)} = \delta_{m_1m_2}P_{m_1}^{(M)}$ and $P_m^{(N)} = \sum_{d\in\mathbb{Z}} P_d^{(M)}\otimes P_{m-d}^{(N-M)}$ (recall that $P_d^{(M)} = 0$ for $d \notin \{0,\dots,M\}$), and without loss of generality singling out the first M variables,

$$\begin{aligned}\Big(\big(P_j^{(M)}h^{(M)}P_k^{(M)}\big)\otimes\mathbb{1}^{\otimes N-M}\Big)P_n^{(N)} &= \Big(\big(P_j^{(M)}h^{(M)}P_k^{(M)}\big)\otimes\mathbb{1}^{\otimes N-M}\Big)\Big(\sum_{d\in\mathbb{Z}}P_d^{(M)}\otimes P_{n-d}^{(N-M)}\Big)\\ &= \big(P_j^{(M)}h^{(M)}P_k^{(M)}\big)\otimes P_{n-k}^{(N-M)}\\ &= \Big(\sum_{d\in\mathbb{Z}}P_d^{(M)}\otimes P_{n-k+j-d}^{(N-M)}\Big)\Big(\big(P_j^{(M)}h^{(M)}P_k^{(M)}\big)\otimes\mathbb{1}^{\otimes N-M}\Big)\\ &= P_{n-k+j}^{(N)}\Big(\big(P_j^{(M)}h^{(M)}P_k^{(M)}\big)\otimes\mathbb{1}^{\otimes N-M}\Big)\,.\end{aligned}$$

It follows from the spectral decomposition (28) of $\mathrm{d}\Gamma(q)$ that

$$\begin{aligned}\mathrm{d}\Gamma^{(M)}\big(P_j^{(M)}h^{(M)}P_k^{(M)}\big)\mathrm{d}\Gamma(q) &= \mathrm{d}\Gamma^{(M)}\big(P_j^{(M)}h^{(M)}P_k^{(M)}\big)\sum_{n\in\mathbb{Z}}n\,P_n^{(N)}\\ &= \sum_{n\in\mathbb{Z}}n\,P_{n-k+j}^{(N)}\mathrm{d}\Gamma^{(M)}\big(P_j^{(M)}h^{(M)}P_k^{(M)}\big)\\ &= \sum_{n\in\mathbb{Z}}(n+k-j)\,P_n^{(N)}\mathrm{d}\Gamma^{(M)}\big(P_j^{(M)}h^{(M)}P_k^{(M)}\big)\\ &= \widehat{\tau_{j-k}Id}\,\mathrm{d}\Gamma^{(M)}\big(P_j^{(M)}h^{(M)}P_k^{(M)}\big)\,,\end{aligned}$$

which, as discussed above, implies the result. □

Proof of Proposition 3.9. Without loss of generality we assume that $\Psi_0 \in \mathfrak{H}_f^{(N)}\cap\mathrm{H}^1(\mathbb{R}^3)^{\otimes N}$ (see Remark 2.7). Using the evolution equation for $\hat{g}$ from Lemma 3.10, we find

$$\begin{aligned}\frac{dS_g}{dt}(\rho_t,p_t) &= \mathrm{Tr}\,\big[-i[H,\rho_t]\hat{g}+\rho_t\,(-i[\mathrm{d}\Gamma(h_{HF}^{(1)}(p_t)),\hat{g}])\big]\\ &= \mathrm{Tr}\,\big[i[H-\mathrm{d}\Gamma(h_{HF}^{(1)}(p_t)),\hat{g}]\rho_t\big]\\ &= \lambda\,\mathrm{Tr}\,\big[i[\frac{1}{2}\mathrm{d}\Gamma^{(2)}(v^{(2)})-\mathrm{d}\Gamma(v_{HF}^{(1)}(p_t)),\hat{g}]\rho_t\big]\end{aligned}$$

with $v_{HF}^{(1)}(p_t) := \mathrm{Tr}_2[v^{(2)}(1-\mathfrak{X})(\mathbb{1}\otimes p_t)]$. Now, recall that $\sum_{m=0}^{M}P_m^{(M)} = \mathbb{1}_{\mathfrak{h}^{\otimes M}}$. Inserting this identity for $M=1$ and $M=2$ and using Lemma 3.11 gives

$$\begin{aligned}\frac{dS_g}{dt}(\rho_t,p_t) &= \frac{\lambda}{2}\,\mathrm{Tr}\,\big[i[\mathrm{d}\Gamma^{(2)}\big(\big(P_0^{(2)}+P_1^{(2)}+P_2^{(2)}\big)v^{(2)}\big(P_0^{(2)}+P_1^{(2)}+P_2^{(2)}\big)\big)\\ &\quad -2\mathrm{d}\Gamma\big(\big(P_0^{(1)}+P_1^{(1)}\big)v_{HF}^{(1)}(p_t)\big(P_0^{(1)}+P_1^{(1)}\big)\big),\hat{g}]\rho_t\big]\\ &= \frac{\lambda}{2}\,\mathrm{Tr}\,\big[i[\mathrm{d}\Gamma^{(2)}(P_0^{(2)}v^{(2)}P_2^{(2)}+P_2^{(2)}v^{(2)}P_0^{(2)}\\ &\quad +P_0^{(2)}v^{(2)}P_1^{(2)}+P_1^{(2)}v^{(2)}P_0^{(2)}\\ &\quad +P_1^{(2)}v^{(2)}P_2^{(2)}+P_2^{(2)}v^{(2)}P_1^{(2)})\end{aligned}$$

$$
\begin{aligned}
&- 2\mathrm{d}\Gamma\big(P_0^{(1)} v_{HF}^{(1)}(p_t) P_1^{(1)} + P_1^{(1)} v_{HF}^{(1)}(p_t) P_0^{(1)}\big), \hat{g}] \rho_t \big] \\
&= \lambda \Im \operatorname{Tr}\left[\big(\mathrm{d}\Gamma^{(2)}(P_1^{(2)} v^{(2)} P_0^{(2)}) - 2\mathrm{d}\Gamma\big(P_1^{(1)} v_{HF}^{(1)}(p_t) P_0^{(1)}\big)\big)\, \rho_t^{[-1,1]} \right] \\
&\quad + \lambda \Im \operatorname{Tr}\left[\mathrm{d}\Gamma^{(2)}(P_2^{(2)} v^{(2)} P_0^{(2)})\, \rho_t^{[-2,2]} \right] \\
&\quad + \lambda \Im \operatorname{Tr}\left[\mathrm{d}\Gamma^{(2)}(P_2^{(2)} v^{(2)} P_1^{(2)})\, \rho_t^{[-1,1]} \right] .
\end{aligned}
$$

We then insert the Fefferman–de la Llave decomposition and Eq. (46) in the first term to get

$$
\begin{aligned}
&\Im \operatorname{Tr}[(\mathrm{d}\Gamma^{(2)}(P_1^{(2)} v^{(2)} P_0^{(2)}) - 2\mathrm{d}\Gamma(P_1^{(1)} v_{HF}^{(1)}(p_t) P_0^{(1)}))\rho_t^{[-1,1]}] \\
&= 2\Im \int \operatorname{Tr}\left[\big(\mathrm{d}\Gamma^{(2)}(q_t X_\omega p_t \otimes p_t X_\omega p_t) \right. \\
&\qquad \left. - \operatorname{Tr}[X_\omega p_t] \mathrm{d}\Gamma(q_t X_\omega p_t) + \mathrm{d}\Gamma(q_t X_\omega p_t X_\omega p_t)\big) \rho_t^{[-1,1]} \right] d\mu(\omega) \\
&= 2\Im \int \operatorname{Tr}\left[\mathrm{d}\Gamma(q_t X_\omega p_t)\big(\mathrm{d}\Gamma(p_t X_\omega p_t) - \operatorname{Tr}[X_\omega p_t]\big)\big) \rho_t^{[-1,1]} \right] d\mu(\omega) ,
\end{aligned}
$$

where we used $P_0^{(1)} = p_t$, $P_1^{(1)} = q_t$, $P_0^{(2)} = p_t^{\otimes 2}$, $P_1^{(2)} = q_t \otimes p_t + p_t \otimes q_t$. □

3.3. Auxiliary lemmas

We prove here three lemmas that we frequently need for estimating the terms $\mathcal{A}_t$, $\mathcal{B}_t$ and $\mathcal{C}_t$ from Proposition 3.9.

For fermionic systems the following bound on $\mathrm{d}\Gamma(A)$ is well-known. Note that this is the only point at which the Fermi statistics enter our paper.

Lemma 3.12. *Let A be a trace-class and self-adjoint operator on a separable Hilbert space $\mathfrak{h}$. Then, as quadratic forms on $\mathfrak{H}_f^{(N)}$,*

$$
\mathrm{d}\Gamma(A) \;\leq\; \|A\|_{\mathcal{L}^1} .
$$

Proof. We use the spectral decomposition $A = \sum_j \lambda_j |\varphi_j\rangle\langle\varphi_j|$ with $\lambda_j \in \mathbb{R}$, $\sum_j |\lambda_j| < \infty$, for some orthonormal basis $(\varphi_j)_{j=1}^\infty$, and we write any vector $\Psi \in \mathfrak{H}_f^{(N)}$ as

$$
\Psi = \sum_{j_1 < \cdots < j_N} \alpha_{j_1,\ldots,j_N} \varphi_{j_1} \wedge \cdots \wedge \varphi_{j_N} ,
$$

where $\|\Psi\|^2 = \sum |\alpha_{j_1,\ldots,j_N}|^2 < \infty$. Then

$$
\begin{aligned}
\langle \Psi, \mathrm{d}\Gamma(A)\Psi \rangle &= \langle \Psi, \sum_{j_1 < \cdots < j_N} (\lambda_{j_1} + \cdots + \lambda_{j_N}) \alpha_{j_1,\ldots,j_N} \varphi_{j_1} \wedge \cdots \wedge \varphi_{j_N} \rangle \\
&= \sum_{j_1 < \cdots < j_N} (\lambda_{j_1} + \cdots + \lambda_{j_N}) |\alpha_{j_1,\ldots,j_N}|^2 \\
&\leq \sum_{j_1 < \cdots < j_N} \|A\|_{\mathcal{L}^1} \, |\alpha_{j_1,\ldots,j_N}|^2 = \|A\|_{\mathcal{L}^1} \, \|\Psi\|^2 ,
\end{aligned}
$$

which yields the result. □

We recall the definition of the direct integral of a family $(B(x))_{x\in\mathbb{R}^3}$ of operators on $\mathfrak{h}$:

$$\Big[\Big(\int_{\mathbb{R}^3}^{\oplus} B(x_1')dx_1'\Big)\psi\Big](x_1,x_2) := \big[\big(\mathbb{1}\otimes B(x_1)\big)\psi\big](x_1,x_2)\,, \tag{49}$$

for any $\psi\in\mathfrak{H}_f^{(2)}$.

Lemma 3.13. *Let A be a bounded non-negative operator on $\mathfrak{h}$ and $(B(x))_{x\in\mathbb{R}^3}$ be a family of non-negative trace class operators on $\mathfrak{h}$. Then*

$$\mathrm{d}\Gamma^{(2)}\Big((\sqrt{A}\otimes\mathbb{1})\int_{\mathbb{R}^3}^{\oplus} B(x_1)dx_1(\sqrt{A}\otimes\mathbb{1})\Big) \leq \mathrm{d}\Gamma\big(\sqrt{A}\ \mathrm{Tr}[B(x)]\ \sqrt{A}\big)\,. \tag{50}$$

If A is also trace-class and such that $\mathrm{Tr}\left[\sqrt{A}\,\mathrm{Tr}[B(x)]\sqrt{A}\right]<\infty$ then

$$\mathrm{d}\Gamma^{(2)}\Big((\sqrt{A}\otimes\mathbb{1})\int_{\mathbb{R}^3}^{\oplus} B(x_1)dx_1(\sqrt{A}\otimes\mathbb{1})\Big) \leq \int A(x;x)\ \mathrm{Tr}[B(x)]dx\,, \tag{51}$$

where $A(x,y)=\sum_{i=1}^{\infty}\lambda_i\varphi_i(x)\overline{\varphi_i(y)}$ denotes the integral kernel of A defined in terms of the spectral decomposition of A.

In particular:

If $B(x)=B$ does not depend on x,

$$\mathrm{d}\Gamma^{(2)}(A\otimes B)\leq \mathrm{Tr}[B]\mathrm{d}\Gamma(A)\,. \tag{52}$$

With $w:\mathbb{R}^3\to\mathbb{R}^+$, $w^{(2)}=w(x_1-x_2)$ and $p=\sum_{i=1}^N|\varphi_i\rangle\langle\varphi_i|$, $\langle\varphi_i|\varphi_j\rangle=\delta_{ij}$ a rank-N projector on $\mathfrak{h}$, we have that

$$\mathrm{d}\Gamma^{(2)}\big((q\otimes p)w^{(2)}(q\otimes p)\big)\leq \mathrm{d}\Gamma\big(q(w*f)q\big)\leq \|w*f\|_\infty\ \mathrm{d}\Gamma(q)\,, \tag{53}$$

$$\mathrm{d}\Gamma^{(2)}\big((p\otimes p)w^{(2)}(p\otimes p)\big)\leq \mathrm{d}\Gamma\big(p(w*f)p\big)\leq \|(w*f)f\|_1\,, \tag{54}$$

where $f(x):=p(x;x):=\sum_{i=1}^N|\varphi_i(x)|^2$ are the diagonal values of the integral kernel of p.

Proof. Let $\Psi^{(N)}\in\mathfrak{H}_f^{(N)}$. With

$$\tilde{\Psi}_{A,x}^{(N-1)}(x_1,\dots,x_{N-1}) := \Big(\big(\mathbb{1}^{\otimes N-1}\otimes\sqrt{A}\big)\Psi^{(N)}\Big)(x_1,\dots,x_{N-1},x)$$

and the direct integral representation we get, using Lemma 3.12,

$$\begin{aligned}
&\Big\langle\Psi^{(N)},\mathrm{d}\Gamma^{(2)}\Big((\sqrt{A}\otimes\mathbb{1})\int_{\mathbb{R}^3}^{\oplus}B(x_1)dx_1(\sqrt{A}\otimes\mathbb{1})\Big)\Psi^{(N)}\Big\rangle\\
&\quad= N(N-1)\int\Big\langle\tilde{\Psi}_{A,x}^{(N-1)},\big(\mathbb{1}^{\otimes N-2}\otimes B(x)\big)\tilde{\Psi}_{A,x}^{(N-1)}\Big\rangle dx\\
&\quad= N\int\Big\langle\tilde{\Psi}_{A,x}^{(N-1)},\mathrm{d}\Gamma\big(B(x)\big)\tilde{\Psi}_{A,x}^{(N-1)}\Big\rangle dx
\end{aligned}$$

$$\leq N \int \left\langle \tilde{\Psi}_{A,x}^{(N-1)}, \mathrm{Tr}[B(x)]\tilde{\Psi}_{A,x}^{(N-1)} \right\rangle dx$$
$$= \left\langle \Psi^{(N)}, \mathrm{d}\Gamma\big(\sqrt{A}\,\mathrm{Tr}[B(x)]\sqrt{A}\big)\Psi^{(N)} \right\rangle,$$

and Eq. (50) follows. If $\mathrm{Tr}[\sqrt{A}\,\mathrm{Tr}[B(x)]\sqrt{A}] < \infty$, then Eq. (51) follows from Lemma 3.12.

The case with $B(x)$ independent of x is clear.

For the second particular case, observe that the operator $(\mathbb{1}\otimes p)w^{(2)}(\mathbb{1}\otimes p)$ can be written as the direct integral

$$(\mathbb{1}\otimes p)w^{(2)}(\mathbb{1}\otimes p) = \int_{\mathbb{R}^3}^{\oplus} p(\tau_{x_1}w)p\,dx_1$$

with $\tau_{x_1}w(x_2) = w(x_2 - x_1)$ a translation of w. Then with $B(x) = p(\tau_x w)p$ and $A = q$ we get Eq. (53), and with $A = p$ we get Eq. (54). □

For $\rho \in \mathcal{L}^1(\mathfrak{H}_f^{(N)})$, let us introduce the shorthand notation

$$\rho^{[j]} := (g - \tau_j g)^{1/2\wedge}\ \rho\ (g - \tau_j g)^{1/2\wedge}, \tag{55}$$
$$\rho^{[-j]} := (\tau_{-j} g - g)^{1/2\wedge}\ \rho\ (\tau_{-j} g - g)^{1/2\wedge}, \tag{56}$$

with $j = 1, 2$, and let $\gamma^{[j]}$ and $\gamma^{[-j]}$ be the corresponding one-particle and $\gamma^{[j](k)}$ and $\gamma^{[-j](k)}$ the corresponding k-particle density matrices (see also Definition 3.2 extended to non-negative and trace class operators whose trace is not necessarily one). Note that $\rho^{[j]}$ and $\rho^{[-j]}$ are not states because their trace is not one, and thus $\gamma^{[j]}$ and $\gamma^{[-j]}$ do not necessarily satisfy Eq. (26).

The next lemma shows the advantage we gain from using the function

$$g_\theta(x) := N^{1-\theta}x1_{[0,N^\theta]}(x) + N1_{(N^\theta,\infty)}(x) \tag{57}$$

in the definition of the degree of evaporation. This lemma is analogous to [50, Lemma 7.1], but note that the use of the functional calculus clarifies the fact that one ultimately uses only inequalities on functions from $\mathbb{R}$ to $\mathbb{R}$.

Lemma 3.14. *For $j \in \{-2, -1, 1, 2\}$, any (normalized) state $\rho \in \mathcal{L}^1\big(\mathfrak{H}_f^{(N)}\big)$, and the function g_θ from (57) (with the notation from (55) and (56)),*

$$\mathrm{Tr}\left[\rho^{[j]}\right] \leq |j|\ N^{1-\theta},$$
$$\mathrm{Tr}\left[\mathrm{d}\Gamma(q)\ \rho^{[j]}\right] \leq |j|\,(|j|+1)\ S_{g_\theta},$$
$$\mathrm{Tr}\left[\mathrm{d}\Gamma^{(2)}(q\otimes q)\ \rho^{[j]}\right] \leq |j|\,(|j|+1)^2\ N^\theta\ S_{g_\theta}.$$

Proof. The inequalities are a direct consequence of the functional calculus, once we observe that $\mathrm{d}\Gamma(q) = \widehat{Id_{\mathbb{R}}}$, $\mathrm{d}\Gamma^{(2)}(q\otimes q) = \mathrm{d}\Gamma(q)^2 - \mathrm{d}\Gamma(q) = (Id_{\mathbb{R}}\cdot(Id_{\mathbb{R}}-1))^\wedge$ and

$$\tau_j g_\theta - \tau_k g_\theta \leq (k-j)\ N^{1-\theta},$$
$$Id_{\mathbb{R}}\cdot(\tau_j g_\theta - \tau_k g_\theta) \leq (k-j)(k-j+1)\ g_\theta,$$
$$Id_{\mathbb{R}}\cdot(Id_{\mathbb{R}}-1)\cdot(\tau_j g_\theta - \tau_k g_\theta) \leq (k-j)\,(k-j+1)^2\ N^\theta\ g_\theta,$$

as inequalities of functions from $\mathbb{R}$ to $\mathbb{R}$, for $-2 \leq j < k \leq 2$. □

3.4. Bound for $\mathcal{A}_t$

Let us first estimate the integrand $\mathcal{A}_t(X_\omega)$ of (41), i.e.,

$$\mathcal{A}_t(X) := 2\Im \operatorname{Tr}\left[\mathrm{d}\Gamma(q_t X p_t)\big(\mathrm{d}\Gamma(p_t X p_t) - \operatorname{Tr}[X p_t]\big)\rho_t^{[-1,1]}\right],$$

where X is an operator on $\mathfrak{h}$ such that $0 \leq X \leq \mathbb{1}$.

Proposition 3.15. *Let X be an operator on $\mathfrak{h}$ such that $0 \leq X \leq \mathbb{1}$ and set $\gamma_t^{[-1]^\perp} := \operatorname{Tr}[\rho_t^{[-1]}] - \gamma_t^{[-1]}$. Then*

$$\mathcal{A}_t(X) \;\leq\; \operatorname{Tr}[p_t X]\,\operatorname{Tr}[X(2q_t\gamma_t^{[1]}q_t + p_t\gamma_t^{[-1]^\perp}p_t)]\,. \tag{58}$$

Proof. Using the Cauchy–Schwarz inequality and $2ab \leq a^2 + b^2$, and then Eq. (46) and Lemma 3.12, we get

$$\begin{aligned}
\mathcal{A}_t(X) \;&=\; 2\Im \operatorname{Tr}\left[\mathrm{d}\Gamma(q_t X p_t)\big(\mathrm{d}\Gamma(p_t X p_t) - \operatorname{Tr}[X p_t]\big)\rho_t^{[-1,1]}\right] \\
&\leq\; \operatorname{Tr}\left[\mathrm{d}\Gamma(q_t X p_t)\,\mathrm{d}\Gamma(p_t X q_t)\,\rho_t^{[1]}\right] \;+\; \operatorname{Tr}\left[\big(\operatorname{Tr}[p_t X] - \mathrm{d}\Gamma(p_t X p_t)\big)^2\rho_t^{[-1]}\right] \\
&\leq\; \operatorname{Tr}\left[\mathrm{d}\Gamma^{(2)}(q_t X p_t \otimes p_t X q_t)\,\rho_t^{[1]}\right] \;+\; \operatorname{Tr}\left[\mathrm{d}\Gamma(q_t X p_t^2 X q_t)\,\rho_t^{[1]}\right] \\
&\quad + \operatorname{Tr}[p_t X]\,\operatorname{Tr}\left[\big(\operatorname{Tr}[p_t X] - \mathrm{d}\Gamma(p_t X p_t)\big)\,\rho_t^{[-1]}\right].
\end{aligned} \tag{59}$$

For the first term on the right-hand side of (59), we apply the Cauchy–Schwarz inequality again and obtain

$$\begin{aligned}
\operatorname{Tr}\left[\mathrm{d}\Gamma^{(2)}(\,q_t X p_t \otimes p_t X q_t)\,\rho_t^{[1]}\right] \;&=\; \operatorname{Tr}\left[\big(q_t\sqrt{X}\otimes p_t\sqrt{X}\big)\,\big(\sqrt{X}p_t\otimes\sqrt{X}q_t\big)\,\gamma_t^{[1](2)}\right] \\
&\leq\; \sqrt{\operatorname{Tr}\left[(q_t X q_t\otimes p_t X p_t)\,\gamma_t^{[1](2)}\right]}\,\sqrt{\operatorname{Tr}\left[(p_t X p_t\otimes q_t X q_t)\,\gamma_t^{[1](2)}\right]} \\
&=\; \operatorname{Tr}\left[(q_t X q_t\otimes p_t X p_t)\,\gamma_t^{[1](2)}\right] \;=\; \operatorname{Tr}\left[\mathrm{d}\Gamma^{(2)}(q_t X q_t\otimes p_t X p_t)\,\rho_t^{[1]}\right].
\end{aligned} \tag{60}$$

Using Lemma 3.13 yields in turn

$$\begin{aligned}
\operatorname{Tr}\left[\mathrm{d}\Gamma^{(2)}(q_t X q_t\otimes p_t X p_t)\,\rho_t^{[1]}\right] \;&\leq\; \operatorname{Tr}\left[p_t^2 X\right]\operatorname{Tr}\left[\mathrm{d}\Gamma(q_t X q_t)\,\rho_t^{[1]}\right] \\
&=\; \operatorname{Tr}[p_t X]\,\operatorname{Tr}\left[X q_t\gamma_t^{[1]}q_t\right].
\end{aligned} \tag{61}$$

For the second term on the right-hand side of (59), we observe that

$$\operatorname{Tr}[\mathrm{d}\Gamma(q_t X p_t^2 X q_t)\,\rho_t^{[1]}] \;=\; \operatorname{Tr}[X p_t^2 X q_t\gamma_t^{[1]}q_t] \;\leq\; \operatorname{Tr}[p_t X]\,\operatorname{Tr}[X q_t\gamma_t^{[1]}q_t]\,,$$

and for the third term on the right-hand side of (59),

$$\operatorname{Tr}[p_t X]\,\operatorname{Tr}\left[\big(\operatorname{Tr}[p_t X] - \mathrm{d}\Gamma(p_t X p_t)\big)\,\rho_t^{[-1]}\right] \;=\; \operatorname{Tr}[p_t X]\,\operatorname{Tr}[(\operatorname{Tr}[\rho_t^{[-1]}]p_t - p_t\gamma_t^{[-1]}p_t)\,X]\,,$$

which yields (58). □

We now give a bound on the integral $\int \mathcal{A}_t(X_\omega)d\mu(\omega)$ using the estimate from Proposition 3.15 on $\mathcal{A}_t(X)$. To get good estimates we take g to be g_θ as in Eq. (37). We use the notation

$$f_{HF}(x) := p_t(x;x) \geq 0\,, \tag{62}$$

where $p_t = \sum_{i=1}^N |\varphi_{i,t}\rangle\langle\varphi_{i,t}|$, with $\langle\varphi_{i,t}|\varphi_{j,t}\rangle = \delta_{ij}$ and $p_t(x;y) := \sum_{i=1}^N \varphi_{i,t}(x)\overline{\varphi_{i,t}(y)}$, which allows us to rewrite the traces as integrals. For example,

$$\mathrm{Tr}[p_t X_{r,z}] \;=\; \int\limits_{|x-z|\leq r} f_{HF}(x)\, d^3x\,.$$

Observe that $\int f_{HF} = N$ and that the quantity $\int f_{HF}^{5/3}$ appearing in Proposition 3.17 is controlled by the Lieb–Thirring inequality, as is discussed in Section 3.7.

Before we give the bound for $\mathcal{A}_t$, let us prove an auxiliary lemma. Let A^c denote the complement of a set A and recall that $B(0,R)$ denotes the ball of radius R centered at 0 in $\mathbb{R}^3$.

Lemma 3.16. *For* $\frac{1}{p_1} + \frac{1}{p_2} + \frac{1}{s} = 2$, *with* $1 \leq p_j, s \leq \infty$, *measurable functions* $\chi, f_1, f_2 : \mathbb{R}^3 \to \mathbb{R}$ *and* $v(x) = |x|^{-1}$,

$$\int (\chi\, v)(x-y)\, f_1(x)\, f_2(y)\, d^3x\, d^3y \;\leq\; \|f_1\|_{p_1}\, \|f_2\|_{p_2}\, \|\chi\, v\|_s\,.$$

Additionally, for $s < 3$,

$$\|1_{B(0,R)}\, v\|_s \;=\; \Big(\frac{4\pi}{3-s}\Big)^{1/s} R^{3/s-1}\,,$$

and, for $s > 3$,

$$\|1_{B(0,R)^c}\, v\|_s \;=\; \Big(\frac{4\pi}{s-3}\Big)^{1/s} R^{3/s-1}\,,$$

with the convention that $\left(\frac{4\pi}{\infty-3}\right)^{1/\infty} := 1$.

Proof. The first relation follows directly from applying Hölder's and Young's inequalities. The second and third relations follow directly from integration. □

With the ingredients above we can give a bound on $\mathcal{A}_t$.

Proposition 3.17. *The estimate*

$$\mathcal{A}_t \leq 5^{-5/6}\, 72\, \pi^{1/3} N^{1/6} \big\|f_{HF}\big\|_{5/3}^{5/6}\, S_{g_\theta} \tag{63}$$

holds.

Proof. By Proposition 3.15,

$$\mathcal{A}_t \leq 2 \int \mathrm{Tr}[p_t X_\omega]\; \mathrm{Tr}[q_t \gamma_t^{[1]} q_t X_\omega]\, d\mu(\omega) + \int \mathrm{Tr}[p_t X_\omega]\; \mathrm{Tr}[p_t \gamma_t^{[-1]^\perp} p_t X_\omega]\, d\mu(\omega)\,. \tag{64}$$

We now explicitly use the Fefferman–de la Llave decomposition of the Coulomb potential. Then, we find that for any non-negative trace-class operator h, using $\int h(y;y)dy = \mathrm{Tr}[h]$, Hölder's inequality and Lemma 3.16 in the end (where we distinguish between the short-range and the long-range part of the potential) gives us that

$$\begin{aligned}\int \mathrm{Tr}[p_t X_\omega]\ \mathrm{Tr}[hX_\omega]\, d\mu(\omega) &= \frac{1}{\pi}\int \mathrm{Tr}[p_t X_{r,z}]\ \mathrm{Tr}[hX_{r,z}]\,\frac{dr}{r^5}\,d^3z\\ &= \frac{1}{\pi}\int \Big(\int_{|x-z|\le r} f_{HF}(x)\,d^3x\Big)\Big(\int_{|y-z|\le r} h(y;y)\,d^3y\Big)\,\frac{dr}{r^5}\,d^3z\\ &= \int \frac{1}{|x-y|} f_{HF}(x)\,h(y;y)\,d^3x\,d^3y\\ &\le \Big(\|1_{B(0,R)}\,v\|_{5/2}\,\|f_{HF}\|_{5/3} + \|1_{B(0,R)^c}\,v\|_\infty\,\|f_{HF}\|_1\Big)\ \mathrm{Tr}[h]\\ &\le \big((8\pi)^{2/5}R^{1/5}\|f_{HF}\|_{5/3} + R^{-1}\|f_{HF}\|_1\big)\ \mathrm{Tr}[h]\,.\end{aligned}$$

Optimizing with respect to $R>0$ yields

$$R \;=\; (8\pi)^{-1/3}5^{5/6}\|f_{HF}\|_1^{5/6}\,\|f_{HF}\|_{5/3}^{-5/6}\,,$$

so that (recall $\|f_{HF}\|_1 = N$)

$$\begin{aligned}\int \mathrm{Tr}[p_t X_\omega]\ \mathrm{Tr}[hX_\omega]\, d\mu(\omega) &\le 5^{-5/6}6(8\pi)^{1/3}\Big(\int f_{HF}^{5/3}\Big)^{1/2}\Big(\int f_{HF}\Big)^{1/6}\ \mathrm{Tr}[h]\\ &= 5^{-5/6}12\pi^{1/3}\,\|f_{HF}\|_{5/3}^{5/6}\,N^{1/6}\ \mathrm{Tr}[h]\,.\end{aligned}$$

We now apply this inequality to (64), i.e., with $h = q_t\gamma_t^{[1]}q_t$ and with $h = p_t\gamma_t^{[-1]^\perp}p_t$. It follows from Lemma 3.14 that

$$\mathrm{Tr}[q_t\gamma_t^{[1]}q_t] = \mathrm{Tr}[q_t\gamma_t^{[1]}] = \mathrm{Tr}[\mathrm{d}\Gamma(q_t)\rho_t^{[1]}] \le 2S_{g_\theta}\,, \tag{65}$$

$$\mathrm{Tr}[p_t\gamma_t^{[-1]^\perp}p_t] = \mathrm{Tr}[p_t]\,\mathrm{Tr}[\rho_t^{[-1]}] - \mathrm{Tr}\left[\mathrm{d}\Gamma(p_t)\rho_t^{[-1]}\right] = \mathrm{Tr}\left[\mathrm{d}\Gamma(q_t)\rho_t^{[-1]}\right] \le 2S_{g_\theta}\,. \tag{66}$$

This proves (63). □

3.5. Bound for $\mathcal{B}_t$

We estimate $\mathcal{B}_t$ in the same fashion as in [50, Lemma 7.3]. Note, that for this term and the $\mathcal{C}_t$ term it is not necessary to use the Fefferman–de la Llave decomposition.

Proposition 3.18. *The estimate*

$$\mathcal{B}_t \le 2^{1/3}\pi^{2/3}\big\|f_{HF}\big\|_{5/3}^{5/6}\,N^{1/6}\big(6S_{g_\theta} + N^{1-\theta}\big) \tag{67}$$

holds.

Proof. We estimate the $\mathcal{B}_t$ term by using the Cauchy–Schwarz inequality to arrive at a three-particle term.

Recall that $\rho_t^{[-2,2]}$ is a rank one operator, i.e., $\rho_t^{[-2,2]} = \big|\Psi^{[-2]}\big\rangle\big\langle\Psi^{[2]}\big|$. For a linear operator A on $\mathfrak{h}$, $j\in\{-2,2\}$ and almost every $x\in\mathbb{R}^3$, we define the vectors $\tilde\Psi_{A,x}^{[j]}\in\mathfrak{H}_f^{(N-1)}$ by

$$\tilde\Psi_{A,x}^{[j]}(x_1,\dots,x_{N-1}) := (\mathbb{1}^{\otimes N-1}\otimes A)\Psi^{[j]}(x_1,\dots,x_{N-1},x)\,.$$

Inserting the form of $v^{(2)}$ as a direct integral into the expression of $\mathcal{B}_t$ in Eq. (42) yields

$$\mathcal{B}_t = \Im\Big\langle \Psi^{[2]}, \mathrm{d}\Gamma^{(2)}\big((q_t\otimes\mathbb{1})\int_{\mathbb{R}^3}^{\oplus} q_t(\tau_{x_1}v)p_t\,dx_1(p_t\otimes\mathbb{1})\big)\Psi^{[-2]}\Big\rangle$$
$$= N\Im\int_{\mathbb{R}^3}\Big\langle \tilde{\Psi}^{[2]}_{q_t,x}, \mathrm{d}\Gamma\big(q_t(\tau_x v)p_t\big)\,\tilde{\Psi}^{[-2]}_{p_t,x}\Big\rangle dx\,.$$

Taking the modulus of both sides and using the Cauchy–Schwarz inequality we obtain

$$\mathcal{B}_t \le \Big(N\int_{\mathbb{R}^3}\big\|\tilde{\Psi}^{[2]}_{q_t,x}\big\|^2 dx\Big)^{1/2}\Big(N\int_{\mathbb{R}^3}\big\|\mathrm{d}\Gamma(q_t(\tau_x v)p_t)\,\tilde{\Psi}^{[-2]}_{p_t,x}\big\|^2 dx\Big)^{1/2}$$
$$= \big\langle\Psi^{[2]},\mathrm{d}\Gamma(q_t)\Psi^{[2]}\big\rangle^{1/2}\Big(N\int_{\mathbb{R}^3}\big\langle\tilde{\Psi}^{[-2]}_{p_t,x},\mathrm{d}\Gamma(p_t(\tau_x v)q_t)\,\mathrm{d}\Gamma(q_t(\tau_x v)p_t)\,\tilde{\Psi}^{[-2]}_{p_t,x}\big\rangle dx\Big)^{1/2}.$$

From Lemma 3.14 we deduce that $\langle\Psi^{[2]},\mathrm{d}\Gamma(q_t)\,\Psi^{[2]}\rangle \le 6S_{g_\theta}$. We estimate the remaining integral using Eq. (46), the Cauchy–Schwarz inequality, Lemma 3.13 and Lemma 3.14:

$$N\int_{\mathbb{R}^3}\Big\langle\tilde{\Psi}^{[-2]}_{p_t,x},\Big[\mathrm{d}\Gamma^{(2)}\big((p_t(\tau_x v)q_t)\otimes(q_t(\tau_x v)p_t)\big)+\mathrm{d}\Gamma\big(p_t(\tau_x v)q_t(\tau_x v)p_t\big)\Big]\tilde{\Psi}^{[-2]}_{p_t,x}\Big\rangle dx$$
$$\le N\int_{\mathbb{R}^3}\Big\langle\tilde{\Psi}^{[-2]}_{p_t,x},\Big[\mathrm{d}\Gamma^{(2)}\big((p_t(\tau_x v)^2p_t)\otimes q_t\big)+\mathrm{d}\Gamma\big(p_t(\tau_x v)^2p_t\big)\Big]\tilde{\Psi}^{[-2]}_{p_t,x}\Big\rangle dx$$
$$= \Big\langle\Psi^{[-2]},\Big[\mathrm{d}\Gamma^{(2)}\big(p_t^{\otimes2}(v^2)^{(2)}p_t^{\otimes2}\big)\mathrm{d}\Gamma(q_t)+\mathrm{d}\Gamma^{(2)}\big(p_t^{\otimes2}(v^2)^{(2)}p_t^{\otimes2}\big)\Big]\Psi^{[-2]}\Big\rangle$$
$$\le \Big(\langle\Psi^{[-2]},\mathrm{d}\Gamma(q_t)\Psi^{[-2]}\rangle+\langle\Psi^{[-2]},\Psi^{[-2]}\rangle\Big)\int_{\mathbb{R}^3}(f_{HF}*v^2)f_{HF}$$
$$\le (6S_{g_\theta}+2N^{1-\theta})\int_{\mathbb{R}^3}(f_{HF}*v^2)f_{HF}\,.$$

By the Hardy–Littlewood–Sobolev inequality (see, e.g., [44, Theorem 4.3]) we find

$$\big\|(v^2*f)f\big\|_1 \le 4^{1/3}\pi^{4/3}\|f\|^2_{3/2}. \tag{68}$$

We then apply Hölder's inequality with $1=\frac{3}{4}+\frac{1}{4}$ to obtain

$$\|f\|^2_{3/2} = \|f^{5/4}f^{1/4}\|_1^{4/3} \le \|f^{5/4}\|_{4/3}^{4/3}\,\|f^{1/4}\|_4^{4/3} = \|f\|_{5/3}^{5/3}\,\|f\|_1^{1/3}\,. \tag{69}$$

Applying this to f_{HF} and $ab\le(a^2+b^2)/2$ to the bound we obtained on $\mathcal{B}_t$ yields the result. □

3.6. Bound for $\mathcal{C}_t$

Our estimate for $\mathcal{C}_t$ is analogous to [50, Lemma 7.3]. Note that for this estimate our choice of the function g_θ is crucial, while in the bounds for $\mathcal{A}_t$ and $\mathcal{B}_t$ we could have used the identity function to obtain the desired estimate. By using g_θ with appropriate $\theta<1$ we obtain the desired N-dependence in the estimate for $\mathcal{C}_t$.

Proposition 3.19. *The estimate*

$$\mathcal{C}_t \le 4\sqrt{2}\,\big\|f_{HF}*v^2\big\|_\infty^{1/2}N^{\theta/2}S_{g_\theta} \tag{70}$$

holds.

Proof. Using the Cauchy–Schwarz inequality, Lemma 3.13 and Lemma 3.14 we find

$$\begin{aligned}\mathcal{C}_t &= 2\Im \operatorname{Tr}\left[\mathrm{d}\Gamma^{(2)}\big((q_t\otimes q_t)v^{(2)}(p_t\otimes q_t)\big)\,\rho_t^{[-1,1]}\right]\\ &\leq 2\Big(\operatorname{Tr}\left[\mathrm{d}\Gamma^{(2)}\big((p_t\otimes q_t)\big(v^{(2)}\big)^2(p_t\otimes q_t)\big)\,\rho_t^{[-1]}\right]\operatorname{Tr}\left[\mathrm{d}\Gamma^{(2)}\big(q_t\otimes q_t\big)\,\rho_t^{[1]}\right]\Big)^{1/2}\\ &\leq 2\Big(\big\|f_{HF}*v^2\big\|_\infty\operatorname{Tr}\left[\mathrm{d}\Gamma\big(q_t\big)\,\rho_t^{[-1]}\right]4N^\theta S_{g_\theta}\Big)^{1/2}\\ &\leq 4\sqrt{2}\,\big\|f_{HF}*v^2\big\|_\infty^{1/2}N^{\theta/2}S_{g_\theta}\,,\end{aligned}$$

which is the result. □

3.7. Kinetic energy estimates and proof of Theorem 3.8

In order to estimate $\|f_{HF}\|_{5/3}^{5/3}$ and $\|f_{HF}*v^2\|_\infty$ in terms of the kinetic energy we use the following inequalities:

Proposition 3.20 *(Lieb–Thirring inequality). (See [46] or [45, p. 73].) Let $\gamma\in\mathcal{L}^1(\mathfrak{h})$ be a one-particle density matrix of finite kinetic energy, i.e., $0\leq\gamma\leq\mathbb{1}$ and $\operatorname{Tr}[-\Delta\gamma]<\infty$. Then*

$$C_{LT}\int f^{5/3}(x)\,d^3x\;\leq\;\operatorname{Tr}[-\Delta\gamma]\,, \tag{71}$$

with $C_{LT}=\frac{9}{5}(2\pi)^{2/3}$ and where $f(x):=\gamma(x;x)$ is the corresponding one-particle density.

Proposition 3.21 *(Hardy's inequality). (See [40] or, e.g., [62].) Let $\gamma\in\mathcal{L}^1(\mathfrak{h})$ be a one-particle density matrix of finite kinetic energy, i.e., $0\leq\gamma\leq\mathbb{1}$ and $\operatorname{Tr}[-\Delta\gamma]<\infty$. Then*

$$\int\frac{f(x)}{|x|^2}\,d^3x\leq 4\,\operatorname{Tr}[-\Delta\gamma]\,, \tag{72}$$

where $f(x):=\gamma(x;x)$ is the corresponding one-particle density.

We now combine the results of Sections 3.2 to 3.6 to prove Theorem 3.8.

Proof of Theorem 3.8. We choose $\theta=\frac{1}{3}$ so that our bound for $\mathcal{C}_t$ is good enough. Collecting the estimates for the $\mathcal{A}_t$, $\mathcal{B}_t$ and $\mathcal{C}_t$ terms from Propositions 3.17, 3.18 and 3.19 and using the kinetic energy inequalities from Propositions 3.20 and 3.21, we can continue the estimate for the time derivative of $S_{g_{1/3},t}$ from Proposition 3.9 and find (recall that $K:=\sup_{t\geq0}\operatorname{Tr}[-\Delta p_t]$)

$$\begin{aligned}\frac{dS_{g_{1/3}}(t)}{dt}&=\lambda\big(\mathcal{A}_t+\mathcal{B}_t+\mathcal{C}_t\big)\\ &\leq\lambda\,5^{-5/6}\,72\,\pi^{1/3}\,N^{1/6}\,\left(\frac{5}{9}(2\pi)^{-2/3}K\right)^{1/2}S_{g_{1/3}}(t)\\ &\quad+\lambda\,2^{1/3}\pi^{2/3}\left(\frac{5}{9}(2\pi)^{-2/3}K\right)^{1/2}N^{1/6}\Big(6S_{g_{1/3}}(t)+N^{2/3}\Big)\\ &\quad+\lambda\,4\sqrt{2}\,(4K)^{1/2}\,N^{1/6}S_{g_{1/3}}(t)\\ &\leq 30\,\lambda\,\sqrt{K}\,N^{1/6}\Big(S_{g_{1/3}}(t)+N^{2/3}\Big)\,.\end{aligned} \tag{73}$$

Integrating this inequality (Grönwall lemma) yields Theorem 3.8. □

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Appendix A. Some results about the theory of the time-dependent Hartree–Fock equation

In this appendix we recall some known facts about the theory of the TDHF equation. We begin by stating a theorem summarizing those results proved in [18] which we use.

Theorem A.1. *Let E a separable Hilbert space, $A : E \supseteq \mathcal{D}(A) \to E$ self-adjoint such that $\exists \mu \in \mathbb{R}$, $A \geq \mu \mathbb{1}$. Let $M := (A - \mu + \mathbb{1})^{1/2}$ and*

$$H^A_{k,p}(E) := \left\{ M^{-k} T M^{-k} \;\middle|\; T = T^*\,, \quad T \in \mathcal{L}^p(E) \right\},$$

equipped with the norm $\|T\|_{k,p,A} = \|M^k T M^k\|_p$, where $\|X\|_p = \mathrm{Tr}[|X|^p]^{1/p}$ for $1 \leq p < \infty$ or $\|X\|_{\mathcal{B}(E)}$ for $p = \infty$ (we write $\mathcal{L}^\infty(E)$ for $\mathcal{B}(E)$). We adopt the special notations $H(E) := H^A_{0,\infty}(E)$ for the space of bounded self-adjoint operators on E and $H^A_1(E) := H^A_{1,1}(E)$ for a weighted space of trace-class operators on E.

Let $\mathcal{W} \in \mathcal{B}(H^A_1(E); H(E))$ be such that

1. $\big(\mathcal{W}(T)M^{-1}\big)(E) \subseteq \mathcal{D}(M)$ *for all* $T \in H^A_1(E)$,
2. $\big(T \mapsto M\mathcal{W}(T)M^{-1}\big) \in \mathcal{B}(H^A_1(E); H(E))$,
3. $\forall T, S \in H^A_1(E)$*:* $\mathrm{Tr}[\mathcal{W}(T)S] = \mathrm{Tr}[\mathcal{W}(S)T]$.

Then

- *For any $T_0 \in H^A_1(E)$ there exists $t_0 > 0$ and $T \in C([0,t_0); H^A_1(E))$ such that, $\forall t \in [0,t_0)$,*

$$T(t) = e^{-itA}\, T_0\, e^{itA} - i \int_0^t e^{-i(t-s)A} \left[\mathcal{W}(T(s)), T(s)\right] e^{i(t-s)A}\, ds\,.$$

  Such a function T is called a local mild solution of the TDHF equation and, provided its interval of definition is maximal, it is unique.
- *If, moreover, $T_0 \in H^A_{2,1}(E)$ then $T \in C^1([0,t_0); H^A_1(E))$ and*

$$\begin{cases} i\frac{dT}{dt}(t) &= \left[A, T(t)\right] \;+\; \left[\mathcal{W}(T(t))\,, T(t)\right], \\ T(0) &= T_0. \end{cases}$$

  Such a function T is called a classical solution of the TDHF equation.

- *Any mild solution to the TDHF equation satisfies*

$$\forall t \in [0, t_0)\,, \quad \mathrm{Tr}\left[MT(t)M\right] + \frac{1}{2}\,\mathrm{Tr}\left[T(t)\,\mathcal{W}(T(t))\right] = \mathrm{Tr}\left[MT_0M\right] + \frac{1}{2}\,\mathrm{Tr}\left[T_0\,\mathcal{W}(T_0)\right].$$

- *If* $\exists k_1 \in \mathbb{R}$ *such that*[1]

$$\left(T \in H_1^A(E)\,,\, 0 \leq T \leq \mathbb{1}\right) \quad \Rightarrow \quad \left(\mathcal{W}(T) \geq k_1\right),$$

and $T_0 \in H_1^A(E)$, $0 \leq T_0 \leq \mathbb{1}$, *then* T *can be extended to the entire positive real axis. Moreover if* $T_0 \in H_A^{2,1}(E)$, *then* T *is the unique global classical solution.*

Remark A.2. In [18] the space $H_{2,1}^A(E)$ is not used. They use a space larger than $H_{2,1}^A(E)$ which is more natural, but less explicit. As it is enough for us to use classical solutions for initial data in $H_{2,1}^A(E)$ and then use a density result, we restrict ourselves to this framework.

We now quote a result which, although not explicitly stated in [18], is a direct consequence of [18] along with [58].

Proposition A.3. *The map*

$$\begin{aligned} H_1^A(E) \times [0,\infty) &\to H_1^A(E) \\ (T_0, t) &\mapsto T(t)\,, \end{aligned}$$

where $T(t)$ *is the (mild) solution to the TDHF equation with initial data* T_0, *is jointly continuous in* T_0 *and* t.

Indeed the proof of existence and uniqueness in [18] is based on the results in [58] which also ensure the continuity with respect to the initial data (see [58, Corollary 1.5, p. 350]).

It was shown in [18] that those results apply to the case $E = \mathfrak{h} = L^2(\mathbb{R}^3)$, $A = -\Delta$,

$$\mathcal{W}(\gamma) \;=\; \mathrm{Tr}_2\left[v^{(2)}(\mathbb{1} - \mathfrak{X})(\mathbb{1} \otimes \gamma)\right],$$

and $v^{(2)} = |x-y|^{-1}$. The proof then extends to the case $A = h^{(1)}$ with $h^{(1)} = -C\Delta + w(x)$, where the external potential w is an infinitesimal perturbation of the Laplacian.

Appendix B. Some estimates of the direct term and the kinetic energy

The dynamics is the free dynamics to leading order in the $\lambda K^{1/2} N^{1/6} t \sim 1$ *regime.* We now substantiate by heuristic argument that, in the particular case of the Coulomb interaction potential, if $\lambda K^{1/2} N^{1/6} t$ is assumed to be of order one, which is the regime where our estimates are relevant, then the evolution is the free evolution to leading order. Note that the exchange term is expected to be subleading with respect to the direct term; we thus neglect the exchange term in the following computation.

We now estimate the effect of the direct term on the time derivative of the average momentum per particle. We denote the Hartree–Fock density at time t by $f_{HF,t} = \sum_{j=1}^N |\varphi_{t,j}|^2$; thus the direct term is the convolution $\lambda v * f_{HF,t}$. For the absolute value of the time derivative of the expectation value of the momentum per particle we find, using $|\nabla v| = 3v^2$ and (68) with (69),

[1] There was a typographical error in Assumption *iv*) in [18], namely, $\mathcal{W}(T)T \geq k_1$ shall be read $\mathcal{W}(T) \geq k_1$.

$$\begin{aligned}\left|N^{-1}\partial_t \operatorname{Tr}\left(p_t(-i\nabla)\right)\right| &= N^{-1}\left|\operatorname{Tr}\left(p_t[h_{HF},\nabla]\right)\right| \\ &\leq \lambda N^{-1}\left|\operatorname{Tr}\left(p_t(\nabla v * f_{HF,t})\right)\right| \\ &\leq 3\lambda N^{-1}\left\|(v^2 * f_{HF,t})f_{HF,t}\right\|_1 \\ &\leq C\lambda N^{-2/3}K. \end{aligned} \tag{B.1}$$

After a time t the effect of the direct term on the momentum is thus expected to be of order $\lambda N^{-2/3}Kt$. Since $\lambda K^{1/2}N^{1/6}t$ is assumed to be of order one, the average change in momentum is of order $K^{1/2}N^{-5/6}$. Since this is much smaller than the average momentum of a particle $(K/N)^{1/2}$, we conclude that the dynamics is, to leading order, free.

The estimate (14) allows to distinguish the free dynamics and the Hartree–Fock dynamics to the next order. Again using heuristic arguments we substantiate that, for large enough kinetic energy $K \gg N^{4/3}$, estimate (14) allows to distinguish the effect of the direct term on the evolution, i.e., our main result shows that the Hartree–Fock equation gives a better approximation to the Schrödinger equation than the free equation. This is because our convergence rate is $N^{-1/6}$ (let us assume $\gamma_0 = p_0$), i.e., for $\lambda K^{1/2}N^{1/6}t$ of order 1, the error between Schrödinger and Hartree–Fock dynamics is for any bounded observable of order $N^{-1/6}$. For $K \gg N^{4/3}$ this rate is much smaller than the average change in momentum estimated above, i.e., $N^{-1/6} \ll K^{1/2}N^{-5/6}$.

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