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Kinetic Energy Estimates for the Accuracy of the Time-Dependent Hartree-Fock Approximation with Coulomb Potential

Volker Bach*, Sébastien Breteaux*, Tim Tzaneteas†

March 6, 2014

Abstract

We study the time evolution of a system of N spinless fermions in \mathbb{R}^3 which interact through a repulsive 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.

I 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,

$$\begin{cases} i\partial_t \Psi_t &= H\Psi_t, \\ \Psi_{t=0} &= \Psi_0. \end{cases} \quad (1)$$

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 paper 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

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given by

$$H = \nu + \sum_{j=1}^N h_j^{(1)} + \lambda \sum_{1 \leq j < k \leq N} v(x_j - x_k). \quad (2)$$

where

- the number $\nu \in \mathbb{R}$ is a constant contribution to the total energy. For example, if we describe a molecule in 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 $N \geq 1$ (although our interest lies ultimately in the description of systems with $N \gg 1$ the estimates in this paper 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 $v(x) := |x|^{-1}$ is the Coulomb potential, for $x \in \mathbb{R}^3 \setminus \{0\}$.

The Hamiltonian specified in (2) describes several situations of interest:

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

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

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 [43, p.21].

Molecule More generally, we can also consider a molecule with $M \in \mathbb{N}$ nuclei of charges $Z_1, \dots, Z_M > 0$ at fixed, distinct positions $R_1, \dots, R_M \in \mathbb{R}^3$ in the Born-Oppenheimer approximation. In this case we have

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

where $\alpha > 0$ is the fine structure constant.

Mean-Field Scaling In the absence of any external potential and relating the large particle number $N \gg 1$ to the small coupling constant $0 < \lambda \ll 1$ in such a way that

$$\nu = 0, \quad h^{(1)} = -\frac{\Delta}{2}, \quad \lambda = \frac{1}{N}. \quad (5)$$

leads to the mean-field model.

Semi-Classical Mean-Field Scaling The semi-classical mean-field scaling combines the mean-field limit of (5) with the semi-classical limit $\hbar \rightarrow 0$ and the semi-classical structure of the initial state (see [15]), i.e.,

$$\nu = 0, \quad h^{(1)} = -\frac{\Delta}{2N^{1/3}}, \quad \lambda = \frac{1}{N^{2/3}}. \quad (6)$$

In the case of the semi-classical mean-field scaling, as presented in [22, 15], our estimate is not very accurate because we do not assume the initial state to possess a specific semi-classical structure.

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,

$$\Phi(x_1, \dots, x_N) = \frac{1}{\sqrt{N!}} \det \begin{pmatrix} \varphi_1(x_1) & \cdots & \varphi_1(x_N) \\ \vdots & \ddots & \vdots \\ \varphi_N(x_1) & \cdots & \varphi_N(x_N) \end{pmatrix}, \quad (7)$$

where the orbitals $\varphi_1, \dots, \varphi_N \in \mathfrak{h}$ are orthonormal. We express (7) 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, 10, 9, 42, 8], i.e., one is interested in finding

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

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 (7). 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

$$i \frac{d\varphi_{t,j}}{dt} = h^{(1)} \varphi_{t,j} + \lambda \sum_{k=1}^N \left\{ [v * |\varphi_{t,k}|^2] \varphi_{t,j} - [v * (\varphi_{t,j} \bar{\varphi}_{t,k})] \varphi_{t,k} \right\}, \quad (8)$$

for $j = 1, \dots, N$.

The TDHF equation (8) can be rewritten in terms of the one-particle density matrix $\eta_t = \sum_{j=1}^N |\varphi_{t,j}\rangle \langle \varphi_{t,j}|$ with $\varphi_{t,j} \in \mathfrak{h}$ and $\langle \varphi_{t,j}, \varphi_{t,k} \rangle = \delta_{j,k}$ as

$$\text{(TDHF)} \quad i \partial_t \eta_t = [h^{(1)}, \eta_t] + \lambda \text{Tr}_2[v^{(2)}, (\eta_t \otimes \eta_t)(1 - \mathfrak{X})]. \quad (9)$$

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. Sometimes, we write $\eta_t^{(2)} = (\eta_t \otimes \eta_t)(1 - \mathfrak{X})$. In the sequel when speaking of the TDHF equation, we refer to (9).

Note that the TDHF equation (9) can be written as $i\partial_t \eta_t = [h_{HF}^{(1)}(\eta_t), \eta_t]$, where the effective HF-Hamiltonian $h_{HF}^{(1)}(\gamma)$ is given by

$$h_{HF}^{(1)}(\gamma) := h^{(1)} + \lambda \text{Tr}_2[v^{(2)}(1_{\mathfrak{h} \otimes \mathfrak{h}} - \mathfrak{X})(1_{\mathfrak{h}} \otimes \gamma)]. \quad (10)$$

Implicitly assuming the existence and regularity of η_t , the HF-Hamiltonian $h_{HF}^{(1)}(\eta_t)$ is self-adjoint with the same domain as $h^{(1)}$, and hence the solution to $\partial_t U_{HF,t} = -i h_{HF}^{(1)}(\eta_t) U_{HF,t}$, with $U_{HF,0} = 1$, is unitary. This has the important consequence that (9) preserves the property of the one-particle density matrix η_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 (9) has been studied in [17] for a bounded two-body interaction. Then the mild solutions of the TDHF equation in the form (8) 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 (9) 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 [56] the existence of mild solutions of the TDHF in the form (8) 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 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 has been proved in [6].

Main Estimate of this Paper (see Theorem II.1) Given a normalized initial state $\Psi_0 \in \mathfrak{H}_f^{(N)} \cap H^1(\mathbb{R}^3)^{\otimes N}$ and the one-particle density matrix $\eta_0 \equiv \eta_{\Phi_{HF,0}}$ associated to 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 $H^1(\mathbb{R}^3)$, the solutions γ_t and η_t to (1) and (9), respectively, obey the trace norm estimate

$$\frac{1}{N} \|\gamma_t - \eta_t\|_{\mathcal{L}^1} \leq \sqrt{\frac{8}{N} \text{Tr}[\gamma_0(1 - \eta_0)]} + \left(56 \lambda N^{1/6} K^{1/2} t\right)^{9/10}$$

where K is a bound on the sum of the kinetic energies of γ_t and η_t which is uniform in time.

Derivation of the TDHF Equation The notion of proximity of two states we use in this paper is defined by expectation values of p -particle observables, where $1 \leq p \ll N$. 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 $\varphi_{t,1}, \dots, \varphi_{t,N}$ are the solutions to (8), then, for any p -particle operator $A^{(p)}$ (i.e., for any bounded operator $A^{(p)}$ on $\mathfrak{h}^{\wedge p} := \mathcal{A}[\mathfrak{h}^{\otimes p}]$), we wish to control the quantity

$$\delta_t^{(p)}(A^{(p)}) := \frac{1}{\|A^{(p)}\|_\infty} \left| \langle \Psi_t, (A^{(p)} \otimes \mathbf{1}_{N-p}) \Psi_t \rangle - \langle \Phi_{HF,t}, (A^{(p)} \otimes \mathbf{1}_{N-p}) \Phi_{HF,t} \rangle \right|.$$

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

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 p -particle density matrix is the trace-class operator $\gamma_\Psi^{(p)}$ on $\mathfrak{H}_f^{(p)}$ whose kernel is given by

$$\begin{aligned} & \gamma_\Psi^{(p)}(x_1, \dots, x_p; y_1, \dots, y_p) \\ &= \frac{N!}{(N-p)!} \int \overline{\Psi(x_1, \dots, x_p, x_{p+1}, \dots, x_N)} \Psi(y_1, \dots, y_p, x_{p+1}, \dots, x_N) d^3x_{p+1} \cdots d^3x_N. \end{aligned}$$

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

$$\delta_t^{(p)}(A^{(p)}) = \frac{1}{\|A^{(p)}\|_\infty} \left| \text{Tr} [(\gamma_{\Psi_t}^{(p)} - \gamma_{\Phi_{HF,t}}^{(p)}) A^{(p)}] \right|$$

and observe that

$$\sup_{A^{(p)} \in \mathcal{B}(\mathfrak{H}_f^{(p)})} \delta_t^{(p)}(A^{(p)}) = \|\gamma_{\Psi_t}^{(p)} - \gamma_{\Phi_{HF,t}}^{(p)}\|_1$$

where $\|\cdot\|_1$ denotes the trace norm. We are thus interested in bounds on $\|\gamma_{\Psi_t}^{(p)} - \gamma_{\Phi_{HF,t}}^{(p)}\|_1$. In the present article we restrict ourselves to the case $p = 1$.

The derivation of the TDHF equations 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. In the case of the dynamics of N identical quantum mechanical particles, the time-dependent Hartree equation, that is the TDHF equation (8) without the exchange term, was first derived rigorously in [54] 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 [40, 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, [51], 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 semiclassical 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 graphs. 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., [54, 13, 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 that solutions to the Gross-Pitaevskii equation. The techniques developed in [47] to study the weakly nonlinear Schrödinger equation are used in [46] 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 commute 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 [49, 41] in the bosonic case, which considerably simplifies the convergence proof for the Hartree equation.

About the fermionic case, the TDHF equation has been derived in [11] in the mean-field 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 [12], still with a bounded two-body potential. In [22], in the semi-classical mean-field scaling, bounds for the Husimi function have been given, assuming the potential to be real-analytic and thus in particular bounded. In the mean-field scaling the TDHF equation has been derived in [32] for the Coulomb potential for a sequences of initial states given as Slater determinants. Up to that point all the method used to derive the TDHF equation had always been based on BBGKY hierarchies. In [15, 14] estimates of $\|\gamma_{N,t} - \eta_{N,t}\|_{\mathcal{L}^1}$ 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 a Grönwall lemma, similarly to [49] 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 time dependent Hartree-Fock equation reduces to the time-dependent Hartree equation.

Sketch of our Derivation of Estimates on the Accuracy of the TDHF Approximation

We derive an estimate on the trace norm of the difference $\gamma_t - \eta_t$ 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 η_t solving the TDHF equation (9). We are inspired by Pickl's method [49], which makes use of a Grönwall lemma for a well-chosen quantity called the *number of bad particles* in [49]. We refer to the quantity we chose to control as the degree, S , of evaporation. In [38, Remark (a) on p. 5] S is called the *degree*

of non-condensation, while in [53] it is called *Verdampfungsgrad*, which translates to *degree of evaporation*.

We show that the degree of evaporation S dominates the square of the trace norm $\|\gamma - \eta\|_{\mathcal{L}^1}$. To obtain the estimates on its time derivative dS/dt 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.) To deal with the Coulomb potential we use 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 [44] then provides an estimate in terms of kinetic energy. Finally, 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.

Discussion of the results Roughly speaking Theorem II.1 below implies that starting from 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 $o((\lambda N^{1/6} K^{1/2})^{-1})$, 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 nearer to a thermodynamic assumption on the system. Another strong point of our result is that it holds in the case of a repulsive Coulomb two-body potential, and the only known previous result with a Coulomb potential was [32].

There, the mean-field scaling was considered and, by a rescaling in time and in space, the result also applies to a large neutral atom (i.e., with charge $N \gg 1$). With the result of [32] the Hartree-Fock approximation is then justified up to times of order $\mathcal{O}(N^{-2})$. Assuming we have a state with a negative energy implies that the kinetic energy is controlled by $\mathcal{O}(N^{7/3})$ (see Sect. II for more details) and our estimate allows us to justify the approximation up to much larger times, of order $o(N^{-4/3})$. Note, however, that our estimate deteriorates if the energy of the state is higher.

A point which could be improved is that our estimate does not take into account any semi-classical structure of the initial data. Hence in the semi-classical mean-field scaling, as in [15] or [22], assuming the kinetic energy to be of order $\mathcal{O}(N^{5/3})$ our result allows only to control the approximation up to times of order $\mathcal{O}(N^{-1/3})$, whereas the estimates in [15] allow to control the approximation up to times of order $\mathcal{O}(1)$ (but only for bounded two-body potentials). Note that our strategy is similar to the one of [15] since we do not use the BBGKY hierarchy but instead make use of a Grönwall lemma (with the same quantity). An important difference is the way to decompose the potential: in [15] a Fourier decomposition is used whereas we use the Fefferman-de la Llave formula.

Outline of the article In Section II we quote our main result, along with applications to molecules or the mean-field limit. In Section III we recall the evolution equation of the one-particle density matrix for the N -particles model. In Section IV

we introduce the degree of evaporation S 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, and prove an estimate of this degree of evaporation S . In Section V we provide estimates which allow us to state our estimate of S in terms of kinetic energy. Appendix A is devoted to the results we use concerning the theory of the TDHF equation.

II Main Result and Applications

Our main result is an estimate 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 (9) in terms of the kinetic energy of the system. As usual, we denote by $H^1(\mathbb{R}^3)$ the sobolev space of weakly differentiable functions with square-integrable derivative.

Theorem II.1. *Specify the following Hypotheses 1–3:*

1. Assume that $\Psi_0 \in \mathfrak{H}_f^{(N)} \cap H^1(\mathbb{R}^3)^{\otimes N}$ is a normalized initial state, and let $\gamma_t := \gamma_{\Psi_t}$ be the one-particle density matrix of $\Psi_t = e^{-iHt}\Psi_0$, [see Eqs. (1) and (2)].
2. Assume that $\Phi_{HF,0} = \varphi_{1,0} \wedge \cdots \wedge \varphi_{N,0}$ is a Slater determinant, with $\varphi_{j,0} \in H^1(\mathbb{R}^3)$ and $\langle \varphi_{j,0}, \varphi_{k,0} \rangle_{\mathfrak{h}} = \delta_{j,k}$, for $1 \leq j, k \leq N$. Let $\eta_0 := \gamma_{\Phi_{HF,0}}$ be the one-particle density matrix of $\Phi_{HF,0}$ and further η_t be the solution to the time-dependent Hartree-Fock equation (9) with initial condition η_0 .
3. Assume that the sum of the kinetic energies of γ_t and η_t is uniformly bounded in time,

$$K := \sup_{t \geq 0} \text{Tr}[(-\Delta)(\gamma_t + \eta_t)] < \infty. \quad (11)$$

Under the assumption of Hypotheses 1–3 the estimate

$$\frac{1}{N} \|\gamma_t - \eta_t\|_{\mathcal{L}^1} \leq \sqrt{\frac{8}{N} \text{Tr}[\gamma_0(1 - \eta_0)]} + \left(56\lambda N^{1/6} K^{1/2} t\right)^{9/10} \quad (12)$$

holds true.

The proof of Theorem II.1 is postponed to Sect VI.

Remark II.2. One of the ingredients of our proof is the Fefferman-de la Llave decomposition of the Coulomb potential [31]

$$\frac{1}{|x|} = \int_0^\infty \frac{16}{\pi r^5} (1_{B(0,r/2)} * 1_{B(0,r/2)})(x) dr, \quad (13)$$

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

$$v(x) = \int_0^\infty g_v(r) (1_{B(0,r/2)} * 1_{B(0,r/2)})(x) dr, \quad (14)$$

with $x \in \mathbb{R}^3 \setminus \{0\}$, was given in [39], and our proof largely generalizes to those potentials v . More precisely, the assertion of Theorem II.1 holds true and without any change in the constants, if we replace the Coulomb potential by any pair potential v that (like $|\cdot|^{-1}$) satisfies Assumption II.4 below. Note that the assumption that v is semi-bounded 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 II.3. *A function $v : \mathbb{R}^3 \setminus \{0\} \rightarrow \mathbb{R}$ satisfies Assumption II.3 if and only if*

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

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 II.4. *(With the same notation as in Assumption II.3.) A function $v : \mathbb{R}^3 \rightarrow \mathbb{R}$ satisfies Assumption II.4 if and only if it satisfies Assumption II.3 along with $|g_v(r)| \leq \frac{16}{\pi} r^{-5}$ and, for some $\mu \in \mathbb{R}$, $v(x) \geq \mu$ for all x .*

In Propositions II.8 and II.9 we give explicit bounds on the kinetic energy K in terms of the energy expectation values $\langle \Psi_0, H \Psi_0 \rangle$ and $\langle \Phi_{HF,0}, H \Phi_{HF,0} \rangle$ of the initial states Ψ_0 and $\Phi_{HF,0}$, respectively, and the ground state energy for the examples presented in Section I. In the case of atoms or molecules this follows from known estimates we now recall.

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

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

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

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

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

Proposition II.5. *Consider a molecule or a neutral atom as in (4) or (3). If $E_{gs} \leq 0$ and*

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

Proof. See [43, p.132]. □

Using Proposition II.5 along with the conservation of the total energy for both the Schrödinger equation and the TDHF equation we get the following bound on the kinetic energy.

Proposition II.6. *Assume that $\Psi_0 \in \mathfrak{H}_f^{(N)} \cap H^1(\mathbb{R}^3)^{\otimes N}$ is normalized and that $\Phi_{HF,0} = \varphi_{1,0} \wedge \cdots \wedge \varphi_{N,0}$ is a Slater determinant, with $\varphi_{j,0} \in H^1(\mathbb{R}^3)$ and $\langle \varphi_{j,0}, \varphi_{k,0} \rangle_{\mathfrak{h}} = \delta_{j,k}$, for $1 \leq j, k \leq N$. In the case of atoms or molecules,*

$$\begin{aligned} K &:= \sup_{t \geq 0} \text{Tr}[(-\Delta)(\gamma_t + \eta_t)] \\ &\leq \left(\sqrt{\mathcal{E}(\Psi_0) - E_{gs}} + \sqrt{-E_{gs}} \right)^2 + \left(\sqrt{\mathcal{E}(\Phi_{HF,0}) - E_{gs}} + \sqrt{-E_{gs}} \right)^2. \end{aligned} \quad (15)$$

Thus, if $\mathcal{E}(\Psi_0) \leq 0$ and $\mathcal{E}(\Phi_{HF,0}) \leq 0$ then

$$K \leq -8E_{gs}. \quad (16)$$

We also recall a known bound for the ground state energy, see [44] or [43], whose units we use.

Proposition II.7 (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*

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

Proposition II.8 (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 II.9 (Mean-field regime without external potential, and non-negative two-body potential). *In the mean-field regime, if $h^{(1)} = -\Delta/2$ and $v(x) \geq 0$ the kinetic energy is bounded by the total energy, which is preserved in time, i.e.,*

$$K \leq \mathcal{E}(\Psi_0) + \mathcal{E}(\Phi_{HF,0}).$$

III Evolution Equation for the One-Particle Density Matrix

For A and $B^{(2)}$ linear operators acting on respectively \mathfrak{h} and $\mathfrak{H}_f^{(2)}$, we use the notation

$$d\Gamma(A) := \sum_{j=1}^N A_j \quad \text{and} \quad d\Gamma^{(2)}(B^{(2)}) := \sum_{\substack{j,k=1 \\ j \neq k}}^N B_{j,k}^{(2)},$$

as operators on $\mathfrak{H}_f^{(N)}$, with A_j acting on the j^{th} factor in $\mathfrak{h}^{\otimes N}$ and $B_{j,k}^{(2)}$ acting on the j^{th} and the k^{th} factor in $\mathfrak{h}^{\otimes N}$, respectively.

Moreover, we use the partial trace $\text{Tr}_2 : \mathcal{L}^1(\mathfrak{H}_f^{(2)}) \rightarrow \mathcal{L}^1(\mathfrak{h})$ which is defined for $B^{(2)} \in \mathcal{L}^1(\mathfrak{H}_f^{(2)})$ to be the operator $\text{Tr}_2[B^{(2)}] \in \mathcal{L}^1(\mathfrak{h})$ such that

$$\text{Tr}[\text{Tr}_2(B^{(2)}) A] = \text{Tr}[B^{(2)} (A \otimes Id_{\mathfrak{h}})], \quad (17)$$

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

Definition III.1. 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 γ_ρ (resp. $\gamma_\rho^{(2)}$), as the operators on \mathfrak{h} (resp. $\mathfrak{H}_f^{(2)}$) such that

$$\forall A \in \mathcal{B}(\mathfrak{h}) : \text{Tr}_{\mathfrak{H}_f^{(N)}}[\rho d\Gamma(A)] = \text{Tr}_{\mathfrak{h}}[\gamma_\rho A], \quad (18)$$

$$\forall B^{(2)} \in \mathcal{B}(\mathfrak{H}_f^{(2)}) : \text{Tr}_{\mathfrak{H}_f^{(N)}}[\rho d\Gamma^{(2)}(B^{(2)})] = \text{Tr}_{\mathfrak{H}_f^{(2)}}[\gamma_\rho^{(2)} B^{(2)}]. \quad (19)$$

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

$$0 \leq \gamma_\rho \leq 1, \quad \text{Tr}_{\mathfrak{h}}[\gamma_\rho] = N, \quad 0 \leq \gamma_\rho^{(2)} \leq N, \quad \text{Tr}_{\mathfrak{H}_f^{(2)}}[\gamma_\rho^{(2)}] = N(N-1).$$

(See [8, Theorem 5.2].) Further note that we are slightly abusing notation since the one-particle density matrix was defined for wave functions, rather than density matrices, before. 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.

Proposition III.2. *If $\Psi_0 \in \mathfrak{H}_f^{(N)} \cap H^1(\mathbb{R}^3)^{\otimes N}$ is normalized then the one- and two-particle density matrices $\gamma_t := \gamma_{\rho_t}$ and $\gamma_t^{(2)} := \gamma_{\rho_t}^{(2)}$, respectively, of $\rho_t := e^{-itH} |\Psi_0\rangle\langle\Psi_0| e^{itH}$ satisfy*

$$i\partial_t \gamma_t = [h^{(1)}, \gamma_t] + \lambda \text{Tr}_2([v^{(2)}, \gamma_t^{(2)}]), \quad (20)$$

where $v^{(2)}$ is the multiplication operator by $v(x-y)$ on a suitable domain containing $\mathfrak{H}_f^{(2)} \cap H^1(\mathbb{R}^3)^{\otimes 2}$.

Proof. Note first that the Hamiltonian H in (2) can be rewritten in *second-quantization* as

$$H = \nu + d\Gamma(h^{(1)}) + \frac{\lambda}{2} d\Gamma^{(2)}(v^{(2)}). \quad (21)$$

Hence, for $A \in \mathcal{B}(\mathfrak{h})$, using (18) and (1), (21) along with the cyclicity of the trace yields

$$\begin{aligned} i\partial_t \text{Tr}[\gamma_t A] &= i\partial_t \text{Tr}[\rho_t d\Gamma(A)] \\ &= \text{Tr}\left(\left[d\Gamma(h^{(1)}) + \frac{\lambda}{2} d\Gamma^{(2)}(v^{(2)}), \rho_t\right] d\Gamma(A)\right) \\ &= \text{Tr}\left(\left[d\Gamma(A), d\Gamma(h^{(1)})\right] \rho_t\right) + \text{Tr}\left(\left[d\Gamma(A), \frac{\lambda}{2} d\Gamma^{(2)}(v^{(2)})\right] \rho_t\right). \end{aligned}$$

The relations

$$\begin{aligned} [\mathrm{d}\Gamma(A), \mathrm{d}\Gamma(B^{(1)})] &= \mathrm{d}\Gamma([A, B^{(1)}]), \\ [\mathrm{d}\Gamma(A), \mathrm{d}\Gamma^{(2)}(B^{(2)})] &= \mathrm{d}\Gamma^{(2)}([A \otimes Id, B^{(2)}]), \end{aligned}$$

which hold for $B^{(j)} \in \mathcal{L}(\mathfrak{H}_f^{(j)})$, along with (18), (19), and the cyclicity of trace then imply that

$$\begin{aligned} i\partial_t \mathrm{Tr}[\gamma_t A] &= \mathrm{Tr}\left(\mathrm{d}\Gamma([A, h^{(1)}]) \rho_t\right) + \mathrm{Tr}\left(\mathrm{d}\Gamma^{(2)}([A \otimes Id, \lambda v^{(2)}]) \rho_t\right) \\ &= \mathrm{Tr}\left([A, h^{(1)}] \gamma_t\right) + \mathrm{Tr}\left([A \otimes Id, \lambda v^{(2)}] \gamma_t^{(2)}\right) \\ &= \mathrm{Tr}\left([h^{(1)}, \gamma_t] A\right) + \mathrm{Tr}\left([\lambda v^{(2)}, \gamma_t^{(2)}] (A \otimes Id)\right), \end{aligned}$$

which is the result, given the defining property (17) of the partial trace. \square

IV Control on the Degree of Evaporation S

We first introduce the degree of evaporation S [53, 38], which is a function of two one-particle density matrices that resembles the relative entropy of two quantum states (see, e.g., [48]).

Definition IV.1. Let $N \in \mathbb{N}$ and

$$\mathfrak{S}_N := \{\gamma \in \mathcal{L}^1(\mathfrak{h}) \mid 0 \leq \gamma \leq 1, \mathrm{Tr}[\gamma] = N\}.$$

The map $S : \mathfrak{S}_N \times \mathfrak{S}_N \rightarrow \mathbb{R}_0^+$ defined by

$$S(\gamma_1, \gamma_2) := \mathrm{Tr}[\gamma_1 - \gamma_1 \gamma_2]. \quad (22)$$

is called the degree of evaporation of γ_1 relative to γ_2 .

Proposition IV.2. For $\gamma_1, \gamma_2 \in \mathfrak{S}_N$, the degree $S(\gamma_1, \gamma_2)$ of evaporation has the following properties

$$0 \leq S(\gamma_1, \gamma_2) \leq N, \quad S(\gamma_1, \gamma_2) = S(\gamma_2, \gamma_1), \quad (23)$$

$$\|\gamma_1 - \gamma_2\|_{\mathcal{L}^2}^2 \leq 2S(\gamma_1, \gamma_2), \quad (24)$$

If furthermore $\gamma_2^2 = \gamma_2$ is a rank- N orthogonal projection then $S(\gamma_2, \gamma_2) = 0$ and

$$\frac{1}{N} \|\gamma_1 - \gamma_2\|_{\mathcal{L}^1} \leq \sqrt{\frac{8}{N} S(\gamma_1, \gamma_2)} \leq \sqrt{\frac{8}{N}} \|\gamma_1 - \gamma_2\|_{\mathcal{L}^1}. \quad (25)$$

Proof. The assertions in (23) are trivial, and (24) follows from

$$\begin{aligned} \|\gamma_1 - \gamma_2\|_{\mathcal{L}^2}^2 &= \mathrm{Tr}[(\gamma_1 - \gamma_2)^2] = \mathrm{Tr}[\gamma_1^2 + \gamma_2^2 - 2\gamma_1 \gamma_2] \\ &= 2S(\gamma_1, \gamma_2) - S(\gamma_1, \gamma_1) - S(\gamma_2, \gamma_2) \leq 2S(\gamma_1, \gamma_2). \end{aligned}$$

For the proof of (25), we first remark that $\gamma_1 - \gamma_2$ has at most N negative eigenvalues (counting multiplicities). This is a well-known consequence of $\gamma_1 - \gamma_2 \geq -\gamma_2$ and the fact that γ_2 is a rank- N orthogonal projection (see, e.g., [50]), but we include its proof for the sake of completeness: Suppose that $\gamma_1 - \gamma_2$ has at least $N + 1$ negative eigenvalues. Then there is a subspace W of dimension $N + 1$ such that $\langle \varphi | (\gamma_1 - \gamma_2) \varphi \rangle < 0$, for all $\varphi \in W \setminus \{0\}$. Since $\gamma_1 \geq 0$, this implies that $\langle \varphi | \gamma_2 \varphi \rangle > 0$, 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 γ_2 has precisely N negative eigenvalues, which contradicts the existence of W .

Denoting the number of negative eigenvalues (counting multiplicities) of $\gamma_1 - \gamma_2$ by K , we consequently have that $K \leq N$. Let $\lambda_1, \dots, \lambda_K$ be these K negative eigenvalues of $\gamma_1 - \gamma_2$, and $\lambda_{K+1}, \lambda_{K+2}, \dots$ be the non-negative ones. Since $\text{Tr}[\gamma_1 - \gamma_2] = 0$, it follows that

$$-(\lambda_1 + \dots + \lambda_K) = \sum_{k=K+1}^{\infty} \lambda_k.$$

Using the Cauchy-Schwarz inequality and $K \leq N$, we obtain

$$\begin{aligned} \|\gamma_1 - \gamma_2\|_{\mathcal{L}^1} &= \sum_{k=K+1}^{\infty} \lambda_k - \sum_{k=1}^K \lambda_k = -2 \sum_{k=1}^K \lambda_k \leq 2\sqrt{K} \left(\sum_{k=1}^K \lambda_k^2 \right)^{1/2} \\ &\leq 2\sqrt{N} \left(\sum_{k=1}^{\infty} \lambda_k^2 \right)^{1/2} = 2\sqrt{N} \|\gamma_1 - \gamma_2\|_{\mathcal{L}^2}, \end{aligned}$$

and the first inequality in (25) result follows from (24). To prove the second inequality in (25), we observe that

$$S(\gamma_1, \gamma_2) = \text{Tr}[\gamma_2(1 - \gamma_1)] = \text{Tr}[\gamma_2(\gamma_2 - \gamma_1)\gamma_2] \leq \|\gamma_2 - \gamma_1\|_{\mathcal{L}^1},$$

using again $\gamma_2 = \gamma_2^2$. \square

Remark IV.3. Although we do not use it, it is interesting to note that the degree of evaporation satisfies the following extensivity property. If $A, B \subseteq \mathbb{R}^3$ are disjoint measurable sets, $\gamma_j = 1_A \gamma_j 1_A + 1_B \gamma_j 1_B \in \mathfrak{S}_N$ and $1_A \gamma_j 1_A \in \mathfrak{S}_{N_A}$, $1_B \gamma_j 1_B \in \mathfrak{S}_{N_B}$ for $j = 1, 2$, where $N_A + N_B = N$, then a direct computation shows that

$$S(1_A \gamma_1 1_A, 1_A \gamma_2 1_A) + S(1_B \gamma_1 1_B, 1_B \gamma_2 1_B) = S(\gamma_1, \gamma_2).$$

The main result of this section is:

Theorem IV.4. *Assume Hypotheses 1 and 2 of Theorem II.1 and that*

$$K_{TF} := \sup_{t \geq 0} \left\{ \int f_{HF,t}^{5/3}, \int f_{T,t}^{5/3} \right\} < \infty, \quad (26)$$

where $f_{HF,t}(x) = \eta_t(x; x)$ and $f_{T,t}(x) = ((1 - \eta_t)\gamma_t(1 - \eta_t))(x; x)$. Then

$$\left(\frac{1}{N} S(\gamma_t, \eta_t) \right)^{5/9} \leq \left(\frac{1}{N} S(\gamma_0, \eta_0) \right)^{5/9} + 25\lambda N^{1/6} K_{TF}^{1/2} t. \quad (27)$$

Theorem IV.4 will be proved in the next subsections. The strategy is to obtain an estimate of dS_t/dt in terms of S_t and then integrate it, in the spirit of a Grönwall lemma.

Remark IV.5. We may evaluate the kernels of η_t and $(1-\eta_t)\gamma_t(1-\eta_t)$ on the diagonal as functions defined almost everywhere since the corresponding operators are trace class.

Remark IV.6. Note that for γ_t , the one-particle density matrix of Ψ_t , and η_t , the one-particle density matrix of the Slater determinant $\Phi_{HF,t}$, the quantity $S(\gamma_t, \eta_t)$ coincides (up to normalization and scaling) with the quantity $\langle \mathcal{U}_N(t; 0)\xi, (\mathcal{N}+1)^k \mathcal{U}_N(t; 0)\xi \rangle$ in [15] in case $\xi = \Omega$ and $k = 1$.

IV.1 Time-Derivative of the Degree of Evaporation

The goal of this and the following section is to estimate the time-derivative dS_t/dt of the degree $S_t := S(\gamma_t, \eta_t)$ of evaporation. To this end, we recall the Fefferman-de la Llave decomposition

$$\frac{1}{|x-y|} = \int_{\mathbb{R}^3} d^3z \int_0^\infty \frac{dr}{\pi r^5} X_{r,z}(x) X_{r,z}(y), \quad (28)$$

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 about $z \in \mathbb{R}^3$. This formula can also be written as

$$v^{(2)} = \int d\mu(\omega) X_\omega \otimes X_\omega, \quad (29)$$

where $\omega = (r, z) \in \mathbb{R}^+ \times \mathbb{R}^3$ and we denote $\int d\mu(\omega) f(\omega) := \int_{\mathbb{R}^3} d^3z \int_0^\infty \frac{dr}{\pi r^5} f(r, z)$. The form (28) is convenient for the estimates derived below, but we note that it agrees with (14), of course.

Proposition IV.7. *The time-derivative of $S_t = S(\gamma_t, \eta_t)$ is*

$$\frac{dS_t}{dt} = \lambda \int \{a_t(X_\omega) + b_t(X_\omega) + c_t(X_\omega)\} d\mu(\omega), \quad (30)$$

where for a linear operator X on \mathfrak{h} such that $0 \leq X \leq 1$, we denote

$$a_t(X) := 2\mathfrak{S}\mathrm{Tr} \left[\rho_t d\Gamma(\eta_t^\perp X \eta_t) (d\Gamma(\eta_t X \eta_t) - \mathrm{Tr}[X \eta_t]) \right], \quad (31)$$

$$b_t(X) := 2\mathfrak{S}\mathrm{Tr} \left[\gamma_t^{(2)} (\eta_t^\perp X \eta_t \otimes \eta_t^\perp X \eta_t) \right], \quad (32)$$

$$c_t(X) := 2\mathfrak{S}\mathrm{Tr} \left[\gamma_t^{(2)} (\eta_t^\perp X \eta_t \otimes \eta_t^\perp X \eta_t^\perp) \right], \quad (33)$$

where $\eta_t^\perp := 1 - \eta_t$.

Before we turn to the proof we note that

$$\eta_t^\perp \eta_t = \eta_t \eta_t^\perp = 0 \quad \text{and} \quad \eta_t^{(2)} = (1 - \mathfrak{X})(\eta_t \otimes \eta_t), \quad (34)$$

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

$$d\Gamma(A) d\Gamma(B) = d\Gamma^{(2)}(A \otimes B) + d\Gamma(AB). \quad (35)$$

Proof. Using the Fefferman-de la Llave decomposition (29) and further (20) and (9) for the derivatives of γ_t and η_t , we first obtain

$$\begin{aligned} \frac{dS_t}{dt} &= i\mathrm{Tr}\left[i\frac{d\gamma_t}{dt}\eta_t + i\gamma_t\frac{d\eta_t}{dt}\right] \\ &= i\mathrm{Tr}\left([h^{(1)}, \gamma_t]\eta_t + \lambda\mathrm{Tr}_2([v^{(2)}, \gamma_t^{(2)}])\eta_t + \gamma_t[h^{(1)}, \eta_t] + \gamma_t\lambda\mathrm{Tr}_2([v^{(2)}, \eta_t^{(2)}])\right] \\ &= i\mathrm{Tr}\left[\lambda\mathrm{Tr}_2[v^{(2)}, \gamma_t^{(2)}]\eta_t + \gamma_t\lambda\mathrm{Tr}_2[v^{(2)}, \eta_t^{(2)}]\right] \end{aligned} \quad (36)$$

$$= \lambda \int 2\Im\mathrm{Tr}[\gamma_t^{(2)}(X_\omega \otimes X_\omega)(\eta_t \otimes Id) + (\gamma_t \otimes Id)\eta_t^{(2)}(X_\omega \otimes X_\omega)] d\mu(\omega) \quad (37)$$

$$= \lambda \int 2\Im\mathrm{Tr}[\gamma_t^{(2)}(X_\omega\eta_t \otimes X_\omega) + (X_\omega\gamma_t \otimes X_\omega)\eta_t^{(2)}] d\mu(\omega), \quad (38)$$

where the equality of (36) and (37) is justified because η_t is a finite-rank operator, and the insertion of $\eta_t = \sum_{j=1}^N |\varphi_{j,t}\rangle\langle\varphi_{j,t}|$ together with Lebesgue's dominated convergence theorem gives this equality, indeed.

For an operator X on \mathfrak{h} such that $0 \leq X \leq 1$ we focus on the integrand in (38). Replacing $\eta_t^{(2)}$ by its explicit form $(1 - \mathfrak{X})(\eta_t \otimes \eta_t)$ and using (35) we have that

$$\begin{aligned} I_t(X) &:= 2\Im\{\mathrm{Tr}[(X\gamma_t \otimes X)\eta_t^{(2)}] + \mathrm{Tr}[\gamma_t^{(2)}(X\eta_t \otimes X)]\} \\ &= 2\Im\{\mathrm{Tr}[(X\gamma_t \otimes X)(1 - \mathfrak{X})(\eta_t \otimes \eta_t)] + \mathrm{Tr}[\rho_t d\Gamma^{(2)}(X\eta_t \otimes X)]\} \\ &= 2\Im\{\mathrm{Tr}[X\gamma_t \eta_t] \mathrm{Tr}[X\eta_t] - \mathrm{Tr}[X\gamma_t \eta_t X\eta_t] \\ &\quad + \mathrm{Tr}[\rho_t d\Gamma^{(2)}(X\eta_t \otimes X\eta_t)] + \mathrm{Tr}[\rho_t d\Gamma^{(2)}(X\eta_t \otimes X\eta_t^\perp)]\} \\ &= 2\Im\{\mathrm{Tr}[X\gamma_t \eta_t] \mathrm{Tr}[X\eta_t] - \mathrm{Tr}[X\gamma_t \eta_t X\eta_t] \\ &\quad + \mathrm{Tr}[\rho_t d\Gamma(X\eta_t) d\Gamma(X\eta_t)] - \mathrm{Tr}[\rho_t d\Gamma(X\eta_t X\eta_t)] \\ &\quad + \mathrm{Tr}[\rho_t d\Gamma^{(2)}(X\eta_t \otimes X\eta_t^\perp)]\}. \end{aligned} \quad (39)$$

Note that $\mathrm{Tr}[\rho_t d\Gamma(X\eta_t X\eta_t)] = \mathrm{Tr}[\gamma_t X\eta_t X\eta_t] = \mathrm{Tr}[\eta_t X\eta_t \gamma_t X] = \overline{\mathrm{Tr}[X\gamma_t \eta_t X\eta_t]}$. Hence, the sum of the second and the fourth term in braces on the right side of (39) is real and does not contribute to $I_t(X)$. This and the definition of the one-particle density matrix γ in (18) yields

$$I_t(X) = 2\Im\{\mathrm{Tr}[\rho_t d\Gamma(X\eta_t) (d\Gamma(X\eta_t) - \mathrm{Tr}[X\eta_t])] + \mathrm{Tr}[\rho_t d\Gamma^{(2)}(X\eta_t \otimes X\eta_t^\perp)]\}.$$

Splitting the identity as $1 = \eta_t + \eta_t^\perp$ and then using again (35) along with (34) gives

$$\begin{aligned} I_t(X) &= 2\Im\{\mathrm{Tr}[\rho_t d\Gamma(X\eta_t) (d\Gamma(\eta_t X\eta_t) - \mathrm{Tr}[X\eta_t])] \\ &\quad + \mathrm{Tr}[\rho_t d\Gamma(X\eta_t) d\Gamma(\eta_t^\perp X\eta_t)] + \mathrm{Tr}[\rho_t d\Gamma^{(2)}(X\eta_t \otimes X\eta_t^\perp)]\} \\ &= 2\Im\{\mathrm{Tr}[\rho_t d\Gamma(\eta_t^\perp X\eta_t) (d\Gamma(\eta_t X\eta_t) - \mathrm{Tr}[X\eta_t])] \\ &\quad + \mathrm{Tr}[\rho_t d\Gamma^{(2)}(X\eta_t \otimes \eta_t^\perp X\eta_t)] + \mathrm{Tr}[\rho_t d\Gamma^{(2)}(X\eta_t \otimes X\eta_t^\perp)]\}. \end{aligned}$$

Using again the same splitting of the identity and then simplifying the terms we obtain

$$\begin{aligned}
I_t(X) &= 2\Im\left\{\mathrm{Tr}\left[\rho_t d\Gamma(\eta_t^\perp X \eta_t) \left(d\Gamma(\eta_t X \eta_t) - \mathrm{Tr}[X \eta_t]\right)\right]\right. \\
&\quad + \mathrm{Tr}\left[\gamma_t^{(2)}(\eta_t X \eta_t \otimes \eta_t^\perp X \eta_t)\right] + \mathrm{Tr}\left[\gamma_t^{(2)}(\eta_t^\perp X \eta_t \otimes \eta_t^\perp X \eta_t)\right] \\
&\quad + \mathrm{Tr}\left[\gamma_t^{(2)}(\eta_t X \eta_t \otimes \eta_t X \eta_t^\perp)\right] + \mathrm{Tr}\left[\gamma_t^{(2)}(\eta_t X \eta_t \otimes \eta_t^\perp X \eta_t^\perp)\right] \\
&\quad + \mathrm{Tr}\left[\gamma_t^{(2)}(\eta_t^\perp X \eta_t \otimes \eta_t X \eta_t^\perp)\right] + \mathrm{Tr}\left[\gamma_t^{(2)}(\eta_t^\perp X \eta_t \otimes \eta_t^\perp X \eta_t^\perp)\right]\left.\right\} \\
&= 2\Im\left\{\mathrm{Tr}\left[\rho_t d\Gamma(\eta_t^\perp X \eta_t) \left(d\Gamma(\eta_t X \eta_t) - \mathrm{Tr}[X \eta_t]\right)\right]\right. \\
&\quad \left. + \mathrm{Tr}\left[\gamma_t^{(2)}(\eta_t^\perp X \eta_t \otimes \eta_t^\perp X \eta_t)\right] + \mathrm{Tr}\left[\gamma_t^{(2)}(\eta_t^\perp X \eta_t \otimes \eta_t^\perp X \eta_t^\perp)\right]\right\}
\end{aligned} \tag{40}$$

The term $2\Im\mathrm{Tr}[\gamma_t^{(2)}(\eta_t^\perp X \eta_t \otimes \eta_t X \eta_t^\perp)]$ is zero because the operator $\eta_t^\perp X \eta_t \otimes \eta_t X \eta_t^\perp$ is invariant under taking the adjoint and conjugation by the exchange operator \mathfrak{X} .

Comparing (40) to (31)–(33), we hence obtain $I_t(X) = a_t(X) + b_t(X) + c_t(X)$ and thus

$$\frac{dS_t}{dt} = \lambda \int I_t(X_\omega) d\mu(\omega) = \lambda \int \{a_t(X_\omega) + b_t(X_\omega) + c_t(X_\omega)\} d\mu(\omega),$$

indeed. □

IV.2 Estimates on $a_t(X)$, $b_t(X)$, $c_t(X)$

Proposition IV.8. *Let X be an operator on \mathfrak{h} such that $0 \leq X \leq 1$ and set $X^\perp := 1 - X$ and $\gamma_t^\perp := 1 - \gamma_t$. Then*

$$a_t(X) \leq \mathrm{Tr}[\eta_t X] \mathrm{Tr}[X(2\eta_t^\perp \gamma_t \eta_t^\perp + \eta_t \gamma_t^\perp \eta_t)], \tag{41}$$

$$b_t(X) \leq 2\sqrt{2} \mathrm{Tr}[\eta_t X] \sqrt{\mathrm{Tr}[X \eta_t^\perp \gamma_t \eta_t^\perp]} \left(\sqrt{\mathrm{Tr}[X \eta_t^\perp \gamma_t \eta_t^\perp]} + 1 \right), \tag{42}$$

$$c_t(X) \leq \mathrm{Tr}[X \eta_t^\perp \gamma_t \eta_t^\perp] \sqrt{\mathrm{Tr}[\eta_t X] \mathrm{Tr}[\eta_t X^\perp]}. \tag{43}$$

The proof of Proposition IV.8 makes much use of the the following lemma, whose proof is demonstrated first.

Lemma IV.9. *Let A and B be two operators on a separable Hilbert space \mathfrak{h} , where A is trace-class and self-adjoint, and $B \geq 0$. Then*

$$d\Gamma(A) \leq \mathrm{Tr}[A_+] \leq \|A\|_{\mathcal{L}^1}, \quad d\Gamma^{(2)}(A \otimes B) \leq \mathrm{Tr}[A_+] d\Gamma(B),$$

where $A_+ := \max\{A, 0\}$ denotes the positive part of a self-adjoint operator A .

Proof of Lemma IV.9. Let $\{\varphi_j\}_{j=1}^\infty \subseteq \mathfrak{h}$ be an orthonormal basis of eigenvectors of A with corresponding eigenvalues $\{\lambda_j\}_{j=1}^\infty \subseteq \mathbb{R}$. Then $A = \sum_j \lambda_j |\varphi_j\rangle\langle\varphi_j|$ and $\sum_j |\lambda_j| < \infty$.

We make use of the fermion creation and annihilation operators

$$\{a_j := a(\varphi_j), a_j^*(\varphi_j)\}_{j=1}^\infty \subseteq \mathcal{B}[\mathfrak{F}_f(\mathfrak{h})],$$

which obey the CAR: $\{a_i, a_j\} = \{a_i^*, a_j^*\} = 0$, $\{a_i, a_j^*\} = \delta_{ij}$. In terms of these creation and annihilation operators, we have that

$$d\Gamma(A) = \sum_{j=1}^{\infty} \lambda_j a_j^* a_j \leq \sum_{j=1}^{\infty} [\lambda_j]_+ a_j^* a_j = d\Gamma(A_+) \leq \sum_{j=1}^{\infty} [\lambda_j]_+ = \text{Tr}[A_+], \quad (44)$$

where we use that $0 \leq a_j^* a_j \leq a_j^* a_j + a_j a_j^* = 1$. This gives the first chain of inequalities.

For the derivation of the inequality on $d\Gamma^{(2)}(A \otimes B)$ we observe that, by the positivity of B and (44), we have that

$$\begin{aligned} d\Gamma^{(2)}(A \otimes B) &= \sum_{j,k=1}^{\infty} \langle \varphi_j | B \varphi_k \rangle a_j^* d\Gamma(A) a_k = \sum_{j,k=1}^{\infty} \langle \sqrt{B} \varphi_j | \sqrt{B} \varphi_k \rangle a_j^* d\Gamma(A) a_k \\ &= \sum_{i=1}^{\infty} M_i^* d\Gamma(A) M_i \leq \sum_{i=1}^{\infty} M_i^* \text{Tr}[A_+] M_i = \text{Tr}[A_+] \sum_{i=1}^{\infty} M_i^* M_i \\ &= \text{Tr}[A_+] d\Gamma[B], \end{aligned} \quad (45)$$

where $M_i := \sum_{k=1}^{\infty} \langle \varphi_i | \sqrt{B} \varphi_k \rangle a_k$. Note that interchanging the order of summations can be easily justified by reading (45) as a quadratic form bound and using Lebesgue's monotone convergence theorem. \square

Proof of (41). Using the Cauchy-Schwarz inequality and $2ab \leq a^2 + b^2$, we get

$$\begin{aligned} a_t(X) &= 2\Im \text{Tr} [d\Gamma(\eta_t^\perp X \eta_t) (d\Gamma(\eta_t X \eta_t) - \text{Tr}[\eta_t X]) \rho_t] \\ &\leq \text{Tr} [d\Gamma(\eta_t^\perp X \eta_t) d\Gamma(\eta_t X \eta_t) \rho_t] + \text{Tr} [(d\Gamma(\eta_t X \eta_t) - \text{Tr}[\eta_t X])^2 \rho_t] \\ &\leq \text{Tr} [d\Gamma^{(2)}(\eta_t^\perp X \eta_t \otimes \eta_t X \eta_t) \rho_t] + \text{Tr} [d\Gamma(\eta_t^\perp X \eta_t^2 X \eta_t) \rho_t] \\ &\quad + \text{Tr}[\eta_t X] \text{Tr}[(\text{Tr}[\eta_t X] - d\Gamma(\eta_t X \eta_t)) \rho_t], \end{aligned} \quad (46)$$

where we use Lemma IV.9 for the second inequality. For the first term on the right side of (46), we apply the Cauchy-Schwarz inequality again and obtain

$$\begin{aligned} \text{Tr} [d\Gamma^{(2)}(\eta_t^\perp X \eta_t \otimes \eta_t X \eta_t) \rho_t] &= \text{Tr} \left[(\eta_t^\perp \sqrt{X} \otimes \eta_t \sqrt{X}) (\sqrt{X} \eta_t \otimes \sqrt{X} \eta_t^\perp) \gamma_t^{(2)} \right] \\ &\leq \sqrt{\text{Tr} [(\eta_t^\perp X \eta_t^\perp \otimes \eta_t X \eta_t) \gamma_t^{(2)}]} \sqrt{\text{Tr} [(\eta_t X \eta_t \otimes \eta_t^\perp X \eta_t^\perp) \gamma_t^{(2)}]} \\ &= \text{Tr} [(\eta_t^\perp X \eta_t^\perp \otimes \eta_t X \eta_t) \gamma_t^{(2)}] = \text{Tr} [d\Gamma^{(2)}(\eta_t^\perp X \eta_t^\perp \otimes \eta_t X \eta_t) \rho_t]. \end{aligned} \quad (47)$$

Using Lemma IV.9 again yields in turn

$$\begin{aligned} \text{Tr} [d\Gamma^{(2)}(\eta_t^\perp X \eta_t^\perp \otimes \eta_t X \eta_t) \rho_t] &\leq \text{Tr}[\eta_t^2 X] \text{Tr} [d\Gamma(\eta_t^\perp X \eta_t) \rho_t] \\ &= \text{Tr}[\eta_t X] \text{Tr} [X \eta_t^\perp \gamma_t \eta_t^\perp]. \end{aligned} \quad (48)$$

For the third term on the right side of (46), we observe that

$$\text{Tr}[\eta_t X] \text{Tr}[(\text{Tr}[\eta_t X] - d\Gamma(\eta_t X \eta_t)) \rho_t] = \text{Tr}[\eta_t X] \text{Tr}[(\eta_t - \eta_t \gamma_t \eta_t) X],$$

and for the second term on the right side of (46), we note that

$$\text{Tr} [d\Gamma(\eta_t^\perp X \eta_t^2 X \eta_t) \rho_t] = \text{Tr} [X \eta_t^2 X \eta_t^\perp \gamma_t \eta_t^\perp] \leq \text{Tr}[\eta_t X] \text{Tr} [X \eta_t^\perp \gamma_t \eta_t^\perp],$$

and hence arrive at (41). \square

Proof of (42). Using the Cauchy-Schwarz inequality, we first note that

$$\begin{aligned}
b_t(X) &= 2\Im\mathrm{Tr}[(\eta_t^\perp X \eta_t \otimes \eta_t^\perp X \eta_t) \gamma_t^{(2)}] \\
&= 2\Im\mathrm{Tr}[d\Gamma(\eta_t X \eta_t^\perp)^2 \rho_t] - 2\Im\mathrm{Tr}[d\Gamma(\eta_t X \eta_t^\perp \eta_t X \eta_t^\perp) \rho_t] \\
&\leq 2\sqrt{\mathrm{Tr}[d\Gamma(\eta_t^\perp X \eta_t) d\Gamma(\eta_t X \eta_t^\perp) \rho_t]} \sqrt{\mathrm{Tr}[d\Gamma(\eta_t X \eta_t^\perp) d\Gamma(\eta_t^\perp X \eta_t) \rho_t]}.
\end{aligned} \tag{49}$$

For the first term in the product on the right side of (49), we observe that

$$\begin{aligned}
&\mathrm{Tr}[d\Gamma(\eta_t^\perp X \eta_t) d\Gamma(\eta_t X \eta_t^\perp) \rho_t] \\
&= \mathrm{Tr}[d\Gamma^{(2)}(\eta_t^\perp X \eta_t \otimes \eta_t X \eta_t^\perp) \rho_t] + \mathrm{Tr}[d\Gamma(\eta_t^\perp X \eta_t^2 X \eta_t^\perp) \rho_t] \\
&\leq 2\mathrm{Tr}[\eta_t X] \mathrm{Tr}[X \eta_t^\perp \gamma_t \eta_t^\perp].
\end{aligned} \tag{50}$$

Indeed, the first term in line (50) is already estimated in (47) and (48) by $\mathrm{Tr}[\eta_t X] \mathrm{Tr}[X \eta_t^\perp \gamma_t \eta_t^\perp]$, while the second term in line (50) is clearly smaller than $\mathrm{Tr}[\eta_t X] \mathrm{Tr}[X \eta_t^\perp \gamma_t \eta_t^\perp]$.

The second term in the product on the right side of (49) is estimated as follows,

$$\begin{aligned}
&\mathrm{Tr}[d\Gamma(\eta_t X \eta_t^\perp) d\Gamma(\eta_t^\perp X \eta_t) \rho_t] \\
&= \mathrm{Tr}[d\Gamma^{(2)}(\eta_t X \eta_t^\perp \otimes \eta_t^\perp X \eta_t) \rho_t] + \mathrm{Tr}[d\Gamma(\eta_t X (\eta_t^\perp)^2 X \eta_t) \rho_t] \\
&\leq \mathrm{Tr}[\eta_t X] \mathrm{Tr}[X \eta_t^\perp \gamma_t \eta_t^\perp] + \mathrm{Tr}[X \eta_t^\perp X \eta_t \gamma_t \eta_t] \\
&\leq \mathrm{Tr}[\eta_t X] \mathrm{Tr}[X \eta_t^\perp \gamma_t \eta_t^\perp] + \mathrm{Tr}[X \eta_t].
\end{aligned}$$

Here we use $\eta_t^\perp \leq 1$, $X^2 \leq X$, $\gamma_t \leq 1$ and $\eta_t^2 \leq \eta_t$ to estimate $\mathrm{Tr}[X \eta_t^\perp X \eta_t \gamma_t \eta_t]$. Further using $\sqrt{1+a} \leq 1 + \sqrt{a}$, which holds true for $a \geq 0$, we arrive at the assertion. \square

Proof of (43). We first remark that $(-i)(\eta_t^\perp X \eta_t - \eta_t X \eta_t^\perp) = i[\eta_t, X]$, and hence

$$c_t(X) = 2\Im\mathrm{Tr}[d\Gamma^{(2)}(\eta_t^\perp X \eta_t \otimes \eta_t^\perp X \eta_t^\perp) \rho_t] = \mathrm{Tr}[d\Gamma^{(2)}(i[\eta_t, X] \otimes \eta_t^\perp X \eta_t^\perp) \rho_t],$$

which, after an application of Lemma IV.9, leads to

$$\begin{aligned}
c_t(X) &\leq \mathrm{Tr}[d\Gamma(\eta_t^\perp X \eta_t^\perp) \rho_t] \|[\eta_t, X]\|_{\mathcal{L}^1} \\
&= \mathrm{Tr}[X \eta_t^\perp \gamma_t \eta_t^\perp] \|\eta_t X \eta_t^\perp - \eta_t^\perp X \eta_t\|_{\mathcal{L}^1}.
\end{aligned}$$

Next, we turn to estimating $\|[\eta_t, X]\|_{\mathcal{L}^1}$. We begin by showing that, for a vector $\varphi \in \mathfrak{h}$, $\|i[|\varphi\rangle\langle\varphi|, X]\|_{\mathcal{L}^1} = \|X\varphi\| \|X^\perp\varphi\|$. The cases $X\varphi = 0$ or $X^\perp\varphi = 0$ are trivial. When both those vectors are non-zero, we set

$$\varphi_1 := \frac{X\varphi}{\|X\varphi\|}, \quad \varphi_2 := \frac{X^\perp\varphi}{\|X^\perp\varphi\|}.$$

Then we can express the commutator as

$$i[|\varphi\rangle\langle\varphi|, X] = \|X\varphi\| \|X^\perp\varphi\| i(|\varphi_2\rangle\langle\varphi_1| - |\varphi_1\rangle\langle\varphi_2|).$$

We now diagonalize this commutator explicitly. Let $\psi_1 := (\varphi_1 + i\varphi_2)/\sqrt{2}$ and $\psi_2 := (\varphi_1 - i\varphi_2)/\sqrt{2}$. Then $\varphi_1 = (\psi_1 + \psi_2)/\sqrt{2}$, $\varphi_2 = i(-\psi_1 + \psi_2)/\sqrt{2}$, $\|\psi_j\| = 1$ and

$$i[|\varphi\rangle\langle\varphi|, X] = \frac{1}{2}\|X\varphi\|\|X^\perp\varphi\|(|\psi_2\rangle\langle\psi_2| - |\psi_1\rangle\langle\psi_1|).$$

Hence

$$\|i[|\varphi\rangle\langle\varphi|, X]\|_{\mathcal{L}^1} = \|X\varphi\|\|X^\perp\varphi\|. \quad (51)$$

Now, we are in position to prove that $\|i[\eta_t, X]\|_{\mathcal{L}^1} \leq \sqrt{\text{Tr}[\eta_t X] \text{Tr}[\eta_t X^\perp]}$. We use the decomposition $\gamma_t = \sum_{\varphi \in \mathcal{B}} \lambda_\varphi |\varphi\rangle\langle\varphi|$ for an orthonormal basis \mathcal{B} of \mathfrak{h} consisting of eigenvectors $\varphi \in \mathcal{B}$ of γ_t with corresponding eigenvalues $\lambda_\varphi \in [0, 1]$. Then, from the previous result and using the Cauchy-Schwarz inequality, we infer that

$$\begin{aligned} \|i[\eta_t, X]\|_{\mathcal{L}^1} &\leq \sum_{\varphi \in \mathcal{B}} \lambda_\varphi \|X\varphi\|\|X^\perp\varphi\| \leq \left(\sum_{\varphi \in \mathcal{B}} \lambda_\varphi \|X\varphi\|^2 \right)^{1/2} \left(\sum_{\varphi \in \mathcal{B}} \lambda_\varphi \|X^\perp\varphi\|^2 \right)^{1/2} \\ &= \sqrt{\text{Tr}[\eta_t X] \text{Tr}[\eta_t X^\perp]}. \end{aligned}$$

Inserting this bound into our estimate of $c_t(X)$ yields (43). \square

IV.3 Integration of the estimates of $a_t(X)$, $b_t(X)$, $c_t(X)$

In view of Propositions IV.7 and IV.8, estimating

$$\frac{1}{\pi} \int \{a_t(X_\omega) + b_t(X_\omega) + c_t(X_\omega)\} \frac{dr}{r^5} d^3z \leq 2(1 + \sqrt{2})I_1 + I'_1 + 2\sqrt{2}I_2 + I_3 \quad (52)$$

is sufficient to estimate $|\lambda^{-1} dS/dt|$, where we used the four integrals:

$$I_1 := \frac{1}{\pi} \int \text{Tr}[\eta_t X_{r,z}] \text{Tr}[\eta_t^\perp \gamma_t \eta_t^\perp X_{r,z}] \frac{dr}{r^5} d^3z, \quad (53)$$

$$I'_1 := \frac{1}{\pi} \int \text{Tr}[\eta_t X_{r,z}] \text{Tr}[\eta_t \gamma_t^\perp \eta_t X_{r,z}] \frac{dr}{r^5} d^3z, \quad (54)$$

$$I_2 := \frac{1}{\pi} \int \text{Tr}[\eta_t X_{r,z}] \sqrt{\text{Tr}[\eta_t^\perp \gamma_t \eta_t^\perp X_{r,z}]} \frac{dr}{r^5} d^3z, \quad (55)$$

$$I_3 := \frac{1}{\pi} \int \sqrt{\text{Tr}[\eta_t X_{r,z}] \text{Tr}[\eta_t X_{r,z}^\perp]} \text{Tr}[\eta_t^\perp \gamma_t \eta_t^\perp X_{r,z}] \frac{dr}{r^5} d^3z. \quad (56)$$

The notations

$$f_{HF}(x) := \eta_t(x; x) \geq 0, \quad (57)$$

$$f_T(x) := (\eta_t^\perp \gamma_t \eta_t^\perp)(x; x) \geq 0, \quad (58)$$

$$f'_T(x) := (\eta_t \gamma_t^\perp \eta_t)(x; x) \geq 0, \quad (59)$$

allow us to rewrite the traces as integrals.

Remark IV.10. Note that for a non-negative trace-class operator A on \mathfrak{h} , there exist an orthonormal basis $(f_j)_{j=1}^\infty$ of \mathfrak{h} and $(\lambda_j)_{j=1}^\infty \in \mathbb{R}_+^{\mathbb{N}}$, such that $A = \sum_{j=1}^\infty \lambda_j |f_j\rangle\langle f_j|$ and $\sum_{j=1}^\infty \lambda_j < \infty$. Hence the kernel of A is defined to be $A(x; y) = \sum_{j=1}^\infty \lambda_j f_j(x) \overline{f_j(y)}$ and, in particular, $A(x; x) = \sum_{j=1}^\infty \lambda_j |f_j(x)|^2$. See also [16].

As an example

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

Observe that $\int f_{HF} = N$ and $\int f_T = \int f_T' = S$. The quantities $\int f_{HF}^{5/3}$ and $\int f_T^{5/3}$ appearing in Proposition IV.11 and Theorem IV.4 are controlled by the Lieb-Thirring inequality, as is discussed in Section V.

Proposition IV.11. *The integrals I_1, I_1', I_2 and I_3 are estimated by*

$$I_1 \leq 5 N^{1/6} \|f_{HF}\|_{5/3}^{5/6} S, \quad (60)$$

$$I_1' \leq 5 N^{1/6} \|f_{HF}\|_{5/3}^{5/6} S, \quad (61)$$

$$I_2 \leq 3 N^{1/6} \|f_{HF}\|_{5/3}^{5/6} S^{1/2}, \quad (62)$$

$$I_3 \leq 11 N^{1/6} \|f_{HF}\|_{5/3}^{5/18} \|f_T\|_{5/3}^{5/9} N^{5/9} S^{4/9}. \quad (63)$$

Lemma IV.12. *For $\frac{1}{p} + \frac{1}{q} + \frac{1}{s} = 2$, with $1 \leq p, q, s \leq \infty$, and any measurable function $\chi : \mathbb{R}^3 \rightarrow \mathbb{R}$,*

$$\int (\chi v)(x-y) f_{HF}(x) f_T(y) d^3x d^3y \leq \|f_{HF}\|_p \|f_T\|_q \|\chi v\|_s.$$

If, additionally, $s < 3$, then

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

and, if $s > 3$, then

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

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

Proof. The first relation is an application of Hölder and Young's inequalities. For $s < 3$, we have that

$$\|1_{B(0,R)} v\|_s^s = \int_{|x| \leq R} |x|^{-s} d^3x = 4\pi \int_{r \leq R} r^{2-s} dr = 4\pi \frac{R^{3-s}}{3-s}.$$

The third relation has a similar proof. □

Proof of Estimates (60) and (61) on I_1 and I_1' . Using the Fefferman-de la Llave decomposition of the Coulomb potential, we indeed have that

$$\begin{aligned} I_1 &= \frac{1}{\pi} \int \mathrm{Tr}[\eta_t X_{r,z}] \mathrm{Tr}[\eta_t^\perp \gamma_t \eta_t^\perp X_{r,z}] \frac{dr}{r^5} d^3z \\ &= \frac{1}{\pi} \int \left(\int_{|x-z| \leq r} f_{HF}(x) d^3x \right) \left(\int_{|y-z| \leq r} f_T(y) d^3y \right) \frac{dr}{r^5} d^3z \\ &= \int \frac{1}{|x-y|} f_{HF}(x) f_T(y) d^3x d^3y \end{aligned}$$

We distinguish between the short-range and the long-range part of the potential for the estimate in S :

$$\begin{aligned} I_1 &\leq (\|1_{B(0,R)} v\|_{5/2} \|f_{HF}\|_{5/3} + \|1_{\mathbb{C}B(0,R)} v\|_{\infty} \|f_{HF}\|_1) \|f_T\|_1 \\ &\leq ((8\pi)^{2/5} R^{1/5} \|f_{HF}\|_{5/3} + R^{-1} \|f_{HF}\|_1) S, \end{aligned}$$

using $\|f_T\|_1 = \text{Tr}[\eta_t^\perp \gamma_t \eta_t^\perp] = S(\gamma_t, \eta_t)$. Optimizing with respect to $R > 0$ yields

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

and

$$I_1 \leq \frac{6}{5} 5^{1/6} (8\pi)^{1/3} \left(\int f_{HF}^{5/3} \right)^{1/2} \left(\int f_{HF} \right)^{1/6} S \leq 5 \|f_{HF}\|_{5/3}^{5/6} N^{1/6} S,$$

which is (60). Estimate (61) follows from the same proof replacing f_T by f'_T and using $\int f'_T = S(\gamma_t, \eta_t)$. \square

Proof of Estimate (62) on I_2 . We split the integral I_2 into the parts for large and for small r and use the Cauchy-Schwarz inequality for large values of r ,

$$\begin{aligned} I_2 &:= \frac{1}{\pi} \int \text{Tr}[\eta_t X_{r,z}] \sqrt{\text{Tr}[\eta_t^\perp \gamma_t \eta_t^\perp X_{r,z}]} \frac{dr}{r^5} d^3 z \\ &\leq \frac{1}{\pi} \int_{r \leq R} \text{Tr}[\eta_t X_{r,z}] \sqrt{\text{Tr}[\eta_t^\perp \gamma_t \eta_t^\perp X_{r,z}]} \frac{dr}{r^5} d^3 z \\ &\quad + \left(\frac{1}{\pi} \int_{r \geq R} \text{Tr}[\eta_t X_{r,z}] \text{Tr}[\eta_t^\perp \gamma_t \eta_t^\perp X_{r,z}] \frac{dr}{r^5} d^3 z \right)^{1/2} \left(\frac{1}{\pi} \int_{r \geq R} \text{Tr}[\eta_t X_{r,z}] \frac{dr}{r^5} d^3 z \right)^{1/2}. \end{aligned}$$

For small r , an application of the Cauchy-Schwarz inequality gives

$$\begin{aligned} &\frac{1}{\pi} \int_{r \leq R} \text{Tr}[\eta_t X_{r,z}] \sqrt{\text{Tr}[\eta_t^\perp \gamma_t \eta_t^\perp X_{r,z}]} \frac{dr}{r^5} d^3 z \\ &= \frac{1}{\pi} \int_{|x-z| \leq r \leq R} f_{HF}(x)^{5/6} f_{HF}(x)^{1/6} \sqrt{\text{Tr}[\eta_t^\perp \gamma_t \eta_t^\perp X_{r,z}]} \frac{dr}{r^5} d^3 z d^3 x \\ &\leq \left(\frac{1}{\pi} \int_{|x-z| \leq r \leq R} f_{HF}(x)^{1/3} \text{Tr}[\eta_t^\perp \gamma_t \eta_t^\perp X_{r,z}] \frac{dr}{r^{6.2}} d^3 z d^3 x \right)^{1/2} \\ &\quad \left(\frac{1}{\pi} \int_{|x-z| \leq r \leq R} f_{HF}(x)^{5/3} \frac{dr}{r^{3.8}} d^3 z d^3 x \right)^{1/2} \\ &\leq \left(\frac{1}{\pi} \int_{\max\{|x-z|, |y-z|\} \leq r \leq R} f_{HF}(x)^{1/3} f_T(y) \frac{dr}{r^{6.2}} d^3 z d^3 x d^3 y \right)^{1/2} \\ &\quad \left(\frac{1}{\pi} \int_{|x-z| \leq r \leq R} f_{HF}(x)^{5/3} \frac{dr}{r^{3.8}} d^3 z d^3 x \right)^{1/2}. \end{aligned}$$

As $3.8 < 4$, this implies that

$$\begin{aligned} \frac{1}{\pi} \int_{|x-z| \leq r \leq R} \frac{dr}{r^{3.8}} d^3 z &= \frac{1}{\pi} \int_{r \leq R} \lambda(\{z \in \mathbb{R}^3 : |x-z| \leq r\}) \frac{dr}{r^{3.8}} \\ &= \frac{1}{\pi} \int_0^R \frac{4\pi r^3}{3} \frac{dr}{r^{3.8}} = \frac{20}{3} R^{1/5} \end{aligned}$$

and hence

$$\left(\frac{1}{\pi} \int_{|x-z| \leq r \leq R} f_{HF}(x)^{5/3} \frac{dr}{r^{3.8}} d^3 z d^3 x\right)^{1/2} \leq \sqrt{\frac{20}{3}} \|f_{HF}\|_{5/3}^{5/6} R^{1/10}.$$

We now consider the term with $r^{-6.2}$. Note that if $|x - y| > 2r$ then

$$\{z \in \mathbb{R}^3 : \max(|x - z|, |y - z|) \leq r\} = \emptyset$$

is void. So, we may assume that

$$|x - y| \leq 2r \leq 2R,$$

and then, with $\theta = |x - y|/(2r)$, we have that

$$\begin{aligned} \lambda\left(\{z \in \mathbb{R}^3 : \max(|x - z|, |y - z|) \leq r\}\right) &= 2r^3 \lambda\left(\{z \in \mathbb{R}^3 : z_1 \geq \theta, |z| \leq 1\}\right) \\ &= 2r^3 \int_{\theta}^1 \pi(1 - z_1^2) dz_1 = 2\pi r^3 \left(\frac{2}{3} - \theta + \frac{\theta^3}{3}\right), \end{aligned}$$

where λ denotes the Lebesgue measure in \mathbb{R}^3 . We can thus control the part of the integral involving the variables r and z as follows,

$$\begin{aligned} \frac{1}{\pi} \int_{\max(|x-z|, |y-z|) \leq r \leq R} \frac{dr}{r^{6.4}} d^3 z &\leq \int_{|x-y|/2}^R 2r^3 \left[\frac{2}{3} - \frac{|x-y|}{2r} + \frac{1}{3} \left(\frac{|x-y|}{2r}\right)^3 \right] \frac{dr}{r^{6.2}} \\ &\leq 2 \int_{|x-y|/2}^{\infty} \left[\frac{2}{3} r^{3-6.2} - r^{2-6.2} \frac{|x-y|}{2} + r^{-6.2} \frac{1}{3} \left(\frac{|x-y|}{2}\right)^3 \right] dr \\ &= \frac{2^{3.2}}{|x-y|^{2.2}} \left(\frac{2}{3 \cdot 2.2} - \frac{1}{3.2} + \frac{1}{3 \cdot 5.2} \right) \\ &= \frac{2^{4.2}}{2.2 \cdot 3.2 \cdot 5.2} \frac{1}{|x-y|^{2.2}}. \end{aligned}$$

Denoting the constant $C' = \frac{10^{3.24.2}}{22 \cdot 32 \cdot 52} = \frac{125}{286} \cdot 2^{0.2} \leq 1$, we hence have

$$\begin{aligned} &\frac{1}{\pi} \int_{\max(|x-z|, |y-z|) \leq r \leq R} f_{HF}(x)^{1/3} f_T(y) \frac{dr}{r^{6.2}} d^3 z d^3 x d^3 y \\ &\leq C' \int_{|x-y| \leq 2R} \frac{f_{HF}(x)^{1/3} f_T(y)}{|x-y|^{2.2}} d^3 x d^3 y \\ &\leq C' \|1_{B(0,2R)} v^{2.2}\|_{5/4} \|f_{HF}^{1/3}\|_5 \|f_T\|_1 \\ &= C' \left(\frac{4\pi}{3 - \frac{11}{4}}\right)^{4/5} [(2R)^{3 - \frac{11}{4}}]^{4/5} \left(\int f_{HF}^{5/3}\right)^{1/5} \left(\int f_T\right) \\ &\leq C' (16\pi)^{4/5} 2^{1/5} R^{1/5} \left(\int f_{HF}^{5/3}\right)^{1/5} \left(\int f_T\right), \end{aligned}$$

where we use Lemma IV.12 to derive the second inequality. Note that the finiteness of the integral $\|1_{B(0,2R)} v^{2.2}\|_{5/4}$ is ensured by $6.2 < 6.4$.

We now turn to the terms for r large. Using Young's inequality we obtain

$$\begin{aligned}
\frac{1}{\pi} \int_{r \geq R} \text{Tr}[\eta_t X_{r,z}] \frac{dr}{r^5} d^3 z &= \frac{1}{\pi} \int_{\max(R, |x-z|) \leq r} f_{HF}(x) d^3 x \frac{dr}{r^5} d^3 z \\
&= \frac{1}{4\pi} \int f_{HF}(x) \frac{d^3 x d^3 z}{\max\{R, |x-z|\}^4} = \frac{1}{4\pi} \|\min\{R^{-1}, v\}^4 * f_{HF}\|_1 \\
&\leq \frac{1}{4\pi} \|\min\{R^{-1}, v\}^4\|_1 \|f_{HF}\|_1 = \frac{1}{4\pi} \left(\frac{4\pi}{3} R^3 R^{-4} + \|1_{B(0,R)} v\|_4^4 \right) \|f_{HF}\|_1 \\
&= \frac{4}{3R} \|f_{HF}\|_1.
\end{aligned}$$

Similarly, we have

$$\begin{aligned}
\frac{1}{\pi} \int_{r \geq R} \text{Tr}[\eta_t X_{r,z}] \text{Tr}[\eta_t^\perp \gamma_t \eta_t^\perp X_{r,z}] \frac{dr}{r^5} d^3 z \\
&= \frac{1}{\pi} \int_{\max(R, |x-z|, |y-z|) \leq r} f_{HF}(x) f_T(y) d^3 x d^3 y \frac{dr}{r^5} d^3 z \\
&= \frac{1}{4\pi} \int f_{HF}(x) f_T(y) \frac{d^3 x d^3 y d^3 z}{\max\{R, |x-z|, |y-z|\}^4} \\
&\leq \frac{1}{4\pi} \int f_{HF}(x) f_T(y) \frac{d^3 x d^3 y d^3 z}{\max\{R, |x-z|\}^4} = \frac{1}{4\pi} \|\min\{R^{-1}, v\}^4 * f_{HF}\|_1 \|f_T\|_1 \\
&\leq \frac{4}{3R} \|f_{HF}\|_1 \|f_T\|_1.
\end{aligned}$$

We collect the inequalities derived above and obtain the following estimate on I_2 ,

$$\begin{aligned}
I_2 &\leq \sqrt{\frac{20}{3}} \|f_{HF}\|_{5/3}^{5/6} R^{1/10} \sqrt{C'(16\pi)^{4/5} 2^{1/5} R^{1/5} \|f_{HF}\|_{5/3}^{1/3} \|f_T\|_1} \\
&\quad + \frac{4}{3R} \|f_{HF}\|_1 \|f_T\|_1^{1/2} \\
&\leq S^{1/2} \left(\sqrt{\frac{20}{3}} (16\pi)^{4/5} 2^{1/5} \frac{125}{286} \cdot 2^{1/5} \|f_{HF}\|_{5/3} R^{1/5} + \frac{4N}{3R} \right) \\
&\leq S^{1/2} \left(\frac{2^{14/5} \pi^{2/5} 25}{\sqrt{286}} \|f_{HF}\|_{5/3} R^{1/5} + \frac{4N}{3R} \right).
\end{aligned}$$

Then optimizing $AR^\alpha + BR^{-\beta}$ with respect to R yields $[(\beta B)^\alpha (\alpha A)^\beta]^{1/(\alpha+\beta)}$ and thus

$$\begin{aligned}
I_2 &\leq S^{1/2} \left[\frac{2^{14/5} \pi^{2/5} 5}{\sqrt{286}} \|f_{HF}\|_{5/3} \left(\frac{4N}{3} \right)^{1/5} \right]^{5/6} \\
&\leq S^{1/2} \left(\frac{2^{14/5} \pi^{2/5} 5}{\sqrt{286}} \right)^{5/6} \|f_{HF}\|_{5/3}^{5/6} \left(\frac{4}{3} \right)^{1/6} N^{1/6} \leq 3 S^{1/2} \|f_{HF}\|_{5/3}^{5/6} N^{1/6},
\end{aligned}$$

which is the asserted estimate. \square

Proof of Estimate (63) on I_3 . We use the decomposition $I_3 = I_{3,1} + I_{3,2}$, where

$$I_{3,1} = \frac{1}{\pi} \int_{r \leq R} \sqrt{\text{Tr}[\eta_t X_{r,z}] \text{Tr}[\eta_t X_{r,z}^\perp]} \text{Tr}[\eta_t^\perp \gamma_t \eta_t^\perp X_{r,z}] \frac{dr}{r^5} d^3 z,$$

$$I_{3,2} = \frac{1}{\pi} \int_{r > R} \sqrt{\text{Tr}[\eta_t X_{r,z}] \text{Tr}[\eta_t X_{r,z}^\perp]} \text{Tr}[\eta_t^\perp \gamma_t \eta_t^\perp X_{r,z}] \frac{dr}{r^5} d^3 z.$$

The first integral $I_{3,1}$ can be estimated using the Hardy-Littlewood maximal function $M_f : \mathbb{R}^3 \rightarrow \mathbb{R}_0^+$, which is defined for $f \in L^1(\mathbb{R}^3)$ by

$$M_f(z) := \sup_{r>0} \left\{ \frac{3}{4\pi r^3} \left(\int_{|x-z| \leq r} f(x) d^3 x \right) \right\},$$

and the maximal inequality,

$$\int M_f^p(z) d^3 z \leq \frac{96}{\pi} \frac{p}{p-1} \int |f(z)|^p d^3 z,$$

which holds true for all $p > 1$, (cf [55, p.58]). Here, we choose $f := f_{HF}$ and $p := 5/3$ and obtain

$$\begin{aligned} I_{3,1} &\leq \frac{\sqrt{N}}{\pi} \int_{r \leq R} \sqrt{\text{Tr}[\eta_t X_{r,z}] \text{Tr}[\eta_t^\perp \gamma_t \eta_t^\perp X_{r,z}]} \frac{dr}{r^5} d^3 z \\ &\leq \frac{\sqrt{N}}{\pi} \sqrt{\frac{4\pi}{3}} \int_{|y-z| \leq r \leq R} M_{HF}^{1/2}(z) f_T(y) \frac{dr}{r^{7/2}} d^3 y d^3 z \\ &\leq \sqrt{\frac{4N}{3\pi}} \frac{2}{5} \int M_{HF}^{1/2}(z) f_T(y) \frac{1(|y-z| \leq R)}{|y-z|^{5/2}} d^3 y d^3 z \\ &\leq \sqrt{\frac{4}{3\pi}} \frac{2}{5} \sqrt{N} \|M_{HF}^{1/2}\|_{10/3} \|f_T\|_{5/3} \|1_{|\cdot| \leq R} v^{5/2}\|_{10/11} \\ &\leq \sqrt{\frac{4}{3\pi}} \frac{2}{5} \left(\frac{240}{\pi}\right)^{3/10} \sqrt{N} \|f_{HF}\|_{5/3}^{1/2} \|f_T\|_{5/3} \left(\frac{11\pi}{2}\right)^{11/10} R^{4/5} \\ &\leq \left(\frac{11^{11} \cdot 2^{21} \cdot \pi^3}{3^2 \cdot 5^7}\right)^{1/10} \sqrt{N} \|f_{HF}\|_{5/3}^{1/2} \|f_T\|_{5/3} R^{4/5}. \end{aligned}$$

The second integral is then estimated as

$$\begin{aligned} I_{3,2} &\leq \frac{N}{\pi} \int_{\max\{|y-z|, R\} \leq r} f_T(y) \frac{dr}{r^5} d^3 z d^3 y \leq \frac{N}{\pi} \int f_T(y) \frac{d^3 z d^3 y}{\max\{|y-z|, R\}^4} \\ &\leq \frac{N}{\pi} \|f_T\|_1 \|\min\{R^{-1}, v\}^4\|_1 \leq \frac{N}{\pi} \left(\frac{4\pi}{3} + 4\pi\right) R^{-1} S \leq \frac{16}{3} N S R^{-1}. \end{aligned}$$

Optimizing with respect to R yields

$$\begin{aligned} I_3 &\leq \left[\left(\frac{16}{3} N S\right)^{4/5} \cdot \frac{4}{5} \cdot \left(\frac{11^{11} \cdot 2^{21} \cdot \pi^3}{3^2 \cdot 5^7}\right)^{1/10} \sqrt{N} \|f_{HF}\|_{5/3}^{1/2} \|f_T\|_{5/3} \right]^{5/9} \\ &\leq \left(\frac{16}{3}\right)^{4/9} \left(\frac{4}{5}\right)^{5/9} \left(\frac{11^{11} \cdot 2^{21} \cdot \pi^3}{3^2 \cdot 5^7}\right)^{1/18} N^{13/18} S^{4/9} \|f_{HF}\|_{5/3}^{5/18} \|f_T\|_{5/3}^{5/9} \\ &\leq \left(\frac{11^{11} \cdot 2^{73} \cdot \pi^3}{3^{10} \cdot 5^{17}}\right)^{1/18} N^{13/18} S^{4/9} \|f_{HF}\|_{5/3}^{5/18} \|f_T\|_{5/3}^{5/9} \\ &\leq 11 N^{1/6} N^{5/9} S^{4/9} \|f_{HF}\|_{5/3}^{5/18} \|f_T\|_{5/3}^{5/9}, \end{aligned}$$

which is (63), indeed. \square

IV.4 Proof of Theorem IV.4

We combine the results of Sections IV.1 to IV.3 to prove Theorem IV.4.

Proof of Theorem IV.4. We abbreviate $S_t := S(\gamma_t, \eta_t)$. Thanks to Propositions IV.7, IV.8 and IV.11 along with (52), we obtain the following estimate of the time derivative of S_t ,

$$\begin{aligned} \frac{dS_t}{dt} &\leq \lambda \sqrt{K_{TF}} N^{1/6} \{ (2(1 + \sqrt{2})5 + 5)S_t + 3S_t^{1/2} + 11N^{5/9}S_t^{4/9} \} \\ &\leq \lambda \sqrt{K_{TF}} N^{1/6} \{ 2(1 + \sqrt{2})5 + 5 + 3 + 11 \} N^{5/9} S_t^{4/9} \\ &\leq 45\lambda \sqrt{K_{TF}} N^{1/6} N^{5/9} S_t^{4/9}, \end{aligned} \quad (64)$$

where we used that $S_t \leq N$ and hence $S_t \leq N^{5/9} S_t^{4/9}$ and $S_t^{1/2} \leq N^{1/2} S_t^{1/2} \leq N^{5/9} S_t^{4/9}$. Thus, integrating the inequality

$$\frac{d}{dt} \left[\left(\frac{S_t}{N} \right)^{5/9} \right] = \frac{5}{9} \frac{1}{N^{5/9} S_t^{4/9}} \frac{dS_t}{dt} \leq 25\lambda \sqrt{K_{TF}} N^{1/6}, \quad (65)$$

we arrive at (27). \square

V Kinetic Energy Estimates

In this section we estimate $\int f_{HF}^{5/3}$ and $\int f_T^{5/3}$ in terms of the kinetic energy of the system.

We first recall the Lieb-Thirring inequality [44, 45].

Proposition V.1 (Lieb-Thirring Inequality). *Let $\gamma \in \mathcal{L}^1(\mathfrak{h})$ be a one-particle density matrix of finite kinetic energy, i.e., $0 \leq \gamma \leq 1$ and $\text{Tr}[-\Delta\gamma] < \infty$. Then, with $C_{LT} = \frac{9}{5}(2\pi)^{2/3}$, the following inequality holds true,*

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

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

Proof. See [43, p.73]. \square

Proposition V.2. *Let $\gamma_t, \eta_t \in \mathcal{L}^1(\mathfrak{h})$ be a two one-particle density matrices of finite kinetic energy, i.e., $0 \leq \gamma_t, \eta_t \leq 1$ and $\text{Tr}[-\Delta(\gamma_t + \eta_t)] < \infty$. Set $\gamma_{T,t} := \eta_t^\perp \gamma_t \eta_t^\perp$ and $f_{T,t}(x) = \gamma_{T,t}(x; x)$. Then*

$$\int f_T^{5/3}(x) d^3x \leq \frac{5}{3} (2\pi)^{-2/3} \text{Tr}[(-\Delta)(\gamma + \eta_t)].$$

Remark V.3. Note that with $i\nabla_\varepsilon := i\nabla/(\varepsilon(-\Delta) + 1)$,

$$\text{Tr}[-\Delta\gamma_{T,t}] = \lim_{\varepsilon \rightarrow 0} \text{Tr}[(i\nabla_\varepsilon)^2 \gamma_{T,t}],$$

and the computations below make sense with $i\nabla$ replaced by the bounded operator $i\nabla_\varepsilon$.

Proof. Applying the Lieb-Thirring inequality, we first observe that

$$C_{LT} \int f_{T,t}^{5/3}(x) d^3x \leq \text{Tr}[(-\Delta)\gamma_{T,t}],$$

and it hence suffices to prove the inequality

$$\text{Tr}[(-\Delta)\gamma_T] \leq 3 \text{Tr}[(-\Delta)(\gamma_t + \eta_t)]. \quad (66)$$

The trace on the left hand side can be written as

$$\begin{aligned} \text{Tr}[(-\Delta)\gamma_{T,t}] &= \text{Tr}[(i\nabla)^2 \eta_t^\perp \gamma_t \eta_t^\perp] \\ &= -\text{Tr}(i\nabla[i\nabla, \eta_t] \gamma_t \eta_t^\perp) + \text{Tr}(i\nabla \eta_t^\perp i\nabla \gamma_t \eta_t^\perp) \\ &= -\text{Tr}(i\nabla[i\nabla, \eta_t] \gamma_t \eta_t^\perp) + \text{Tr}(\eta_t^\perp i\nabla \gamma_t [i\nabla, \eta_t]) + \text{Tr}(\eta_t^\perp i\nabla \gamma_t i\nabla \eta_t^\perp). \end{aligned} \quad (67)$$

We estimate the third term on the right side of (67) by using that $\eta_t^\perp \leq 1$ as follows,

$$\begin{aligned} \text{Tr}(\eta_t^\perp i\nabla \gamma_t i\nabla \eta_t^\perp) &= \text{Tr}[(i\nabla \gamma_t^{1/2})^* (\eta_t^\perp)^2 (i\nabla \gamma_t^{1/2})] \\ &\leq \text{Tr}[(i\nabla \gamma_t^{1/2})^* (i\nabla \gamma_t^{1/2})] = \text{Tr}[(-\Delta)\gamma_t]. \end{aligned}$$

For the second term on the right side of (67) we observe that

$$\begin{aligned} |\text{Tr}(\eta_t^\perp i\nabla \gamma_t [i\nabla, \eta_t])| &= |\text{Tr}(\eta_t^\perp i\nabla \gamma_t \eta_t i\nabla)| \\ &\leq \sqrt{\text{Tr}[\eta_t^\perp i\nabla \gamma_t^2 i\nabla \eta_t^\perp] \text{Tr}[i\nabla \eta_t^2 i\nabla]} \\ &\leq \sqrt{\text{Tr}[(-\Delta)\gamma_t] \text{Tr}[(-\Delta)\eta_t]} \leq \frac{1}{2} \text{Tr}[(-\Delta)(\gamma_t + \eta_t)]. \end{aligned}$$

For the first term on the right side of (67) we start with the observation that

$$|\text{Tr}(i\nabla[i\nabla, \eta_t] \gamma_t \eta_t^\perp)| \leq |\text{Tr}([i\nabla, \eta_t] \gamma_t [i\nabla, \eta_t])| + |\text{Tr}([i\nabla, \eta_t] \gamma_t i\nabla \eta_t^\perp)|. \quad (68)$$

The first term in (68) with two commutators is estimated by

$$0 \leq \text{Tr}([i\nabla, \eta_t] \gamma_t [i\nabla, \eta_t]) \leq \text{Tr}([i\nabla, \eta_t]^2) \leq 2\text{Tr}[(-\Delta)\eta_t],$$

while the second term in (68) can be estimated by

$$\begin{aligned} |\text{Tr}([i\nabla, \eta_t] \gamma_t i\nabla \eta_t^\perp)| &\leq \sqrt{\text{Tr}([i\nabla, \eta_t]^2) \text{Tr}(\eta_t^\perp i\nabla \gamma_t^2 i\nabla \eta_t^\perp)} \\ &\leq \sqrt{2 \text{Tr}[(-\Delta)\eta_t] \text{Tr}[(-\Delta)\gamma_t]} \leq \frac{1}{2} \text{Tr}[(-\Delta)(\gamma_t + \eta_t)]. \end{aligned}$$

□

VI Proof of Theorem II.1

We now deduce Theorem II.1 from the results proven in Sect. III to V.

Proof of Theorem II.1. Using Proposition IV.2 and $S(\gamma_t, \eta_t) = \text{Tr}[(1 - \eta_t)\gamma_t]$, we obtain

$$\frac{1}{N} \|\gamma_t - \eta_t\|_{\mathcal{L}^1} \leq \sqrt{8} \left[\left(\frac{1}{N} S(\gamma_t, \eta_t) \right)^{5/9} \right]^{9/10}.$$

Theorem IV.4 then gives a bound on the degree of evaporation $S(\gamma, \eta_t)$

$$\frac{1}{N} \|\gamma_t - \eta_t\|_{\mathcal{L}^1} \leq \sqrt{8} \left[\left(\frac{1}{N} S(\gamma_0, \eta_0) \right)^{5/9} + 25\lambda N^{1/6} K_{TF}^{1/2} t \right]^{9/10}.$$

Propositions V.1 and V.2 allow to bound K_{TF} (see (26) for the definition of K_{TF})

$$\begin{aligned} \frac{1}{N} \|\gamma_t - \eta_t\|_{\mathcal{L}^1} &\leq \sqrt{8} \left(\frac{1}{N} S(\gamma_0, \eta_0) \right)^{1/2} + \left(8^{5/9} \sqrt{\frac{5}{3}} (2\pi)^{-1/3} 25\lambda N^{1/6} K^{1/2} t \right)^{9/10} \\ &\leq \sqrt{8} \left(\frac{1}{N} S(\gamma_0, \eta_0) \right)^{1/2} + \left(56\lambda N^{1/6} K^{1/2} t \right)^{9/10} \end{aligned} \quad (69)$$

where we used $(a+b)^\theta \leq a^\theta + b^\theta$, for $a, b \geq 0$ and $\theta \in [0, 1]$. We thus obtain estimate (12). \square

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 regrouping those of the results proved in [18] which we use.

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

$$H_{k,p}^A(E) := \{ M^{-k} T M^{-k} \mid T = T^*, \quad T \in \mathcal{L}^p(E) \},$$

equipped with the norm $\|T\|_{k,p,A} = \|M^k T M^k\|_p$ where $\|X\|_p = \text{Tr}[|X|^p]^{1/p}$ 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 $H(E) := H_{0,\infty}^A(E)$ for the space of bounded self-adjoint operators on E and $H_1^A(E) := H_{1,1}^A(E)$ for a weighted space of trace-class operators on E .

Let $\mathcal{W} \in \mathcal{B}(H_1^A(E); H(E))$ such that

1. $(\mathcal{W}(T)M^{-1})(E) \subseteq \mathcal{D}(M)$,
2. $(T \mapsto M\mathcal{W}(T)M^{-1}) \in \mathcal{B}(H_1^A(E); H(E))$,
3. $\forall T, S \in H_1^A(E) : \text{Tr}[\mathcal{W}(T)S] = \text{Tr}[\mathcal{W}(S)T]$.

Then

- For any $T_0 \in H_1^A(E)$ there exists $t_0 > 0$ and $T \in C([0, t_0]; H_1^A(E))$ such that, $\forall t \in [0, t_0)$,

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

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 $T_0 \in H_{2,1}^A(E)$ then $T \in C^1([0, t_0]; H_1^A(E))$ and

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

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

- Any mild solution to the TDHF equation satisfies

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

- If $\exists k_1 \in \mathbb{R}$, such that*

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

and $T_0 \in H_1^A(E)$, $0 \leq T_0 \leq 1$ then T can be extended to all the 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 [52].

Proposition A.3. *The application*

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

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 [52] which also ensure the continuity with respect to the initial data (see Corollary 1.5 p.350 in [52]).

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

$$\mathcal{W}(\gamma) = \text{Tr}_2[v^{(2)}(1 - \mathfrak{X})(1 \otimes \gamma)],$$

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.

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

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