Regular Articles

Local existence and uniqueness of solutions to the time-dependent Kohn–Sham equations coupled with classical nuclear dynamics

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A R T I C L E   I N F O

A B S T R A C T

We prove the short-time existence and uniqueness of solutions to the initial-value problem associated with a class of time-dependent Kohn–Sham equations coupled with Newtonian nuclear dynamics, combining Yajima's theory for time-dependent Hamiltonians with Duhamel's principle, based on suitable Lipschitz estimates. We consider a pure power exchange term within a generalisation of the so-called local-density approximation, identifying a range of exponents for the existence and uniqueness of H^2 solutions to the Kohn–Sham equations.

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1. Introduction

1.1. Main result

In this article we study the existence and uniqueness of solutions to the system

$$i\dot{\psi}_k = -\tfrac{1}{2}\Delta_x \psi_k - \sum_{K=1}^{N_{\mathrm{nuc}}} \frac{Z_K}{|\cdot - X_K|}\psi_k + \left(\frac{1}{|\cdot|} * \rho\right)\psi_k + \lambda\rho^{q-1}\psi_k, \tag{1.1a}$$

$$\ddot{X}_K = \frac{Z_K}{M_K}\left[\int \frac{x - X_K}{|x - X_K|^3}\rho(x)\mathrm{d}x + \sum_{L=1,L\neq K}^{N_{\mathrm{nuc}}} Z_L\frac{X_K - X_L}{|X_K - X_L|^3}\right], \tag{1.1b}$$

where $N_{\mathrm{el}}, N_{\mathrm{nuc}} \in \mathbb{N}$, $Z_K \in \mathbb{N}$, $M_K \geq 0$, $\lambda \in \mathbb{R}$ and $q > 1$ are given, and $k = 1, \ldots, N_{\mathrm{el}}$ and $K = 1, \ldots, N_{\mathrm{nuc}}$. We use the short-hand notation $\psi = (\psi_1, \ldots, \psi_{N_{\mathrm{el}}})$ and $X = (X_1, \ldots, X_{N_{\mathrm{nuc}}}) \in \mathbb{R}^{3N_{\mathrm{nuc}}}$, with

$$\psi_k = \psi_k(x,t), \qquad X_K = X_K(t), \qquad x \in \mathbb{R}^3, \qquad t \geq 0.$$

In the above equations, for all $\psi(\cdot) : [0, \tau_{\max}) \to H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$, we set

$$\rho = \sum_{k=1}^{N_{\mathrm{el}}} |\psi_k|^2.$$

Moreover, for all $X \in \mathbb{R}^{3N_{\mathrm{nuc}}}$ and $k = 1, \ldots, N_{\mathrm{el}}$, we define

$$(H[X,\rho]\psi)_k := -\tfrac{1}{2}\Delta_x \psi_k - \sum_{K=1}^{N_{\mathrm{nuc}}} \frac{Z_K}{|\cdot - X_K|}\psi_k + \left(\frac{1}{|\cdot|} * \rho\right)\psi_k + \lambda\rho^{q-1}\psi_k. \tag{1.2}$$

The dynamics of the elements $X(\cdot) : [0, \tau_{\max}) \to \mathbb{R}^{3N_{\mathrm{nuc}}}$ is driven by the acceleration function $A = A^1 + A^2$, whose components are defined as

$$A_K^1[\rho](X) := \frac{Z_K}{M_K}\int \frac{x - X_K}{|x - X_K|^3}\rho(x)\mathrm{d}x, \qquad A_K^2(X) := \frac{Z_K}{M_K}\sum_{L=1,L\neq K}^{N_{\mathrm{nuc}}} Z_L\frac{X_K - X_L}{|X_K - X_L|^3}. \tag{1.3}$$

The main result of this article is as follows.

Theorem 1.1. *Let $q \geq 7/2$ and $\lambda \in \mathbb{R}$. Further, let $\psi^0 \in H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$, $V^0 \in \mathbb{R}^{3N_{\mathrm{nuc}}}$ and $X^0 \in \mathbb{R}^{3N_{\mathrm{nuc}}}$ be given, with $X_K^0 \neq X_L^0$ for $K \neq L$.*
Then there exists $\tau > 0$ such that the initial-value problem associated with the system (1.1) with $\psi(0) = \psi^0$, $X(0) = X^0$ and $\dot{X}(0) = V^0$ has a unique solution $(\psi, X) \in \mathcal{X}(\tau)$, where

$$\mathcal{X}(\tau) := C^1([0,\tau]; L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})) \cap C^0([0,\tau]; H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})) \times C^2([0,\tau]; \mathbb{R}^{3N_{\mathrm{nuc}}}).$$

1.2. Physical motivation

Problems such as (1.1) describe the nonadiabatic dynamics of molecular, spin-unpolarised systems involving an even number $N_{\text{el}} \in 2\mathbb{N}$ of electrons and $N_{\text{nuc}} \in \mathbb{N}$ nuclei with masses $M_1, \ldots, M_{N_{\text{nuc}}}$ and charges $Z_1, \ldots, Z_{N_{\text{nuc}}}$. See, for example, [2,14,17,19–21,26,27,30], which form a sample of the extensive body of literature on both physical and mathematical aspects of the so-called density-functional theory, which comprises the framework of the time-dependent Kohn–Sham (TDKS) equations, given in (1.1a). These equations, which with the use of (1.2) can be written as

$$i\dot{\psi} = H[X, \rho]\psi, \tag{1.4}$$

describe the electronic evolution in terms of single-particle wave functions ψ_k, known in the physical literature as the Kohn–Sham (KS) orbitals. The TDKS equations have been extensively considered as an approximation to the time-dependent Schrödinger equation, which reduces the electronic dynamics to a single-particle description based on the KS density function ρ. For convenience, we briefly recall the physical interpretation of each potential in the KS Hamiltonian H from (1.2), which can be written as

$$H[X, \rho] = -\tfrac{1}{2}\Delta_x + V_{\text{ext}}[X] + V_{\text{Hxc}}[\rho], \qquad V_{\text{Hxc}} := V_{\text{H}} + V_{\text{x}} + V_{\text{c}}. \tag{1.5}$$

The different terms appearing in (1.5) are defined as follows.

The electrostatic potential

$$V_{\text{ext}}[X](x) := -\sum_{K=1}^{N_{\text{nuc}}} \frac{Z_K}{|x - X_K|}$$

is an external potential, generated by the nuclei, which represents the Coulombic nucleus-electron interactions. The Hartree potential

$$V_{\text{H}}[\rho] := |\cdot|^{-1} * \rho$$

corresponds to the Coulombic electron-electron interactions. The remaining term, the exchange-correlation potential $V_{\text{x}} + V_{\text{c}}$, is not explicitly known: in the local-density approximation (LDA) introduced by Kohn and Sham [19], for the exchange potential V_{x} an approximation based on the homogeneous electron gas approximation is chosen [25]. In this article, we study a generalisation of this exchange potential, of the form

$$V_{\text{x}}[\rho] := \lambda \rho^{q-1},$$

where $\lambda \in \mathbb{R}$ and $q > 1$. Hereafter, we set the so-called correlation potential to zero, namely,

$$V_{\text{c}} \equiv 0,$$

and write accordingly $V_{\text{Hxc}} = V_{\text{Hx}}$. In most cases, there is no closed form for the correlation potential, and one has to resort to numerical presentations, which are too complex to investigate in the same manner as we handle the other terms. See, for example, [2,18], where the case $V_{\text{c}} \not\equiv 0$ is considered in time-independent or specific time-dependent settings.

In the coupling of (1.4) with the equations (1.1b) describing the nuclear dynamics, which with the use of (1.3) can be written as

$$\ddot{X} = A[\rho](X), \tag{1.6}$$

we apply the so-called mean-field or Ehrenfest dynamics approach—see, for example, [1, Section V], [15, Section 2.1], [23, Section II.3] and [29]—based on factorising the total wave function into a product of fast (electronic) and slow (nuclear) particle parts. In this nonadiabatic mixed quantum–classical dynamics method, we use a point-nucleus approximation rather than the Born–Oppenheimer approximation, which would assume some requirements for the system under consideration. In this way, we can ignore the quantum nature of the nuclei, since these are much heavier than electrons, and consider them as classical point particles. This mean-field description can be understood as a semiclassical limit of the time-dependent self-consistent field (or Hartree) method, from which the Hamilton–Jacobi equation (equivalent to Newton's law of motion) for the nuclei can be derived. According to this description, the nuclei move subject to a single effective potential of Hellman–Feynman type, corresponding to an average over quantum states:

$$M_K A_K[\rho](X) = -\nabla_{X_K} W[\rho](X) \quad \text{for all } K,$$

where

$$W[\rho](X) := (V_{\text{ext}}[X], \rho)_{L^2(\mathbb{R}^3)} + W_{\text{nn}}(X),$$

$$W_{\text{nn}}(X) := \frac{1}{2} \sum_{K,L=1,K\neq L}^{N_{\text{nuc}}} \frac{Z_K Z_L}{|X_K - X_L|} \tag{1.7}$$

describe the interaction of the electrons with the external potential, and the Coulombic internal nuclear interactions, respectively. Note that the exchange term does not appear in the coupling of (1.4) with (1.1b), as it does not describe electrostatic interaction, but describes interactions between the electrons. Also, we note that our equations (1.1) can be regarded as a Hamiltonian system. The total energy E associated with this system is given by

$$E[X, \psi] := E_{\text{kin}}[X, \psi] + W[|\psi|^2](X) + E_{\text{H}}[|\psi|^2] + E_{\text{x}}[|\psi|^2],$$

where

$$E_{\text{kin}}[X, \psi] := \frac{1}{2} \sum_{K=1}^{N_{\text{nuc}}} M_K |\dot{X}_K|^2 + \frac{1}{2} \sum_{k=1}^{N_{\text{el}}} \int |\nabla_x \psi_k(x)|^2 \mathrm{d}x$$

is the kinetic energy of the system. The other terms are potential energies:

$$E_{\text{H}}[|\psi|^2] := \frac{1}{2} \iint \frac{|\psi(x)|^2 |\psi(x')|^2}{|x - x'|} \mathrm{d}x\, \mathrm{d}x'$$

is the Hartree electrostatic self-repulsion of the KS electron density, and

$$E_{\text{x}}[|\psi|^2] := \frac{\lambda}{q} \int |\psi(x)|^{2q} \mathrm{d}x$$

is the exchange energy, whose functional derivative coincides with the exchange potential V_{x}. The total energy E and $\|\psi\|_{L^2(\mathbb{R}^3; \mathbb{C}^{N_{\text{el}}})}$ are quantities which are conserved under the dynamics, as is customary for Hamiltonian systems.

Cancès and Le Bris [8] considered similar electronic evolution equations coupled with classical nuclear dynamics consistent with the mean-field Ehrenfest approach. They studied a system involving the Hartree–Fock equations:

$$i\dot{\psi}^{\mathrm{HF}} = H^{\mathrm{HF}}[X, \psi^{\mathrm{HF}}]\psi^{\mathrm{HF}}, \tag{1.8a}$$

$$\ddot{X} = A[\rho](X), \tag{1.8b}$$

where $\rho = \sum_{k=1}^{N_{\mathrm{el}}} |\psi_k^{\mathrm{HF}}|^2$, the Hartree–Fock Hamiltonian is defined as

$$H^{\mathrm{HF}}[X, \psi^{\mathrm{HF}}] := -\tfrac{1}{2}\Delta_x + V_{\mathrm{ext}}[X] + V_{\mathrm{H}}[\rho] + V_{\mathrm{x}}^{\mathrm{HF}}[\psi^{\mathrm{HF}}]$$

and

$$(V_{\mathrm{x}}^{\mathrm{HF}}[\psi^{\mathrm{HF}}]\psi^{\mathrm{HF}})_k := -\sum_{\ell=1}^{N_{\mathrm{el}}} (\overline{\psi_\ell^{\mathrm{HF}}}\psi_k^{\mathrm{HF}} * |\cdot|^{-1})\psi_\ell^{\mathrm{HF}}$$

is known as the Hartree–Fock exchange potential. Here, ψ_k^{HF} are single-particle wave functions. In [8], the global-in-time existence and uniqueness of solutions to (1.8) in H^2 are based on the result obtained by Yajima [31] for the existence of propagators associated with linear, time-dependent Hamiltonians. The proof in [8] consists of two main steps: a fixed-point argument to show the existence of short-time solutions, based on Lipschitz estimates in $H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$, and a Grönwall-type argument which relies on energy conservation, conservation of the $L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$ norm of ψ^{HF} and estimates of the solutions ψ^{HF} in the $H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$ norm.

To the best of our knowledge, since the article by Cancès and Le Bris [8], only a few contributions have dealt with the coupling of a system describing electronic evolution with nuclear dynamics; this is the case, for instance, in [3], where existence and regularity questions are studied for a similar system, in the case $\lambda = 0$. Considerable attention has also been devoted to Schrödinger–Poisson-type equations, which include the Hartree–Fock and TDKS equations; see, for instance, [2,5–7,9,10,13,18,22,32]. We also mention [28], where existence, uniqueness and regularity questions are investigated for TDKS equations set on bounded space domains, in relation to control problems. None of the contributions cited above considered the combined nuclear and electronic dynamics as described in our system.

1.3. Article outline

This article is organised as follows.

In Section 2, we recall the relevant results from [31] on the construction and properties of a family of propagators

$$U(t, s) : L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}) \longrightarrow L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}),$$

with $t, s \in [0, \Theta]$, associated with the linear parts of the KS Hamiltonians $H[X(t), \rho]$ for $t \in [0, \Theta]$, with $0 < \Theta < \infty$, and some results from [8] on the bounds on the operator norms of these propagators.

In Sections 3 and 4 we define bounded regions $\mathcal{B}_{\mathrm{el}}(\tau)$ and $\mathcal{B}_{\mathrm{nuc}}(\tau)$, designed to seek solutions to (1.4) and (1.6), respectively, on a time interval $[0, \tau]$, and the mappings

$$\mathcal{N} : \mathcal{B}_{\mathrm{el}}(\tau) \longrightarrow \mathcal{B}_{\mathrm{nuc}}(\tau) \cap C^2([0, \tau]; \mathbb{R}^{3N_{\mathrm{nuc}}}), \qquad \mathcal{E} : \mathcal{B}_{\mathrm{nuc}}(\tau) \longrightarrow \mathcal{B}_{\mathrm{el}}(\tau)$$

which connect these solutions.

In Section 5, in view of a Duhamel-type argument developed in later sections, we state and prove some Lipschitz estimates on the nonlinear mapping

$$\psi \longmapsto V_{\mathrm{Hx}}[|\psi|^2]\psi.$$

The restriction $q \geq 7/2$ arises from these estimates.

Next, we prove in Section 6 that for some $\tau > 0$ and any fixed $\psi \in \mathcal{B}_{\mathrm{el}}(\tau)$, the Cauchy problem (1.6) has a unique solution $X \in \mathcal{B}_{\mathrm{nuc}}(\tau) \cap C^2([0, \tau]; B_\delta(X^0))$, with $B_\delta(X^0)$ denoting a closed ball of radius δ centred at X^0, and the mapping $\mathcal{N}[\psi] = X$ is bounded with respect to the $C^1([0, \tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})$ topology, and continuous as a map from $C^0([0, \tau]; L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))$ to $C^0([0, \tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})$. We construct these solutions as fixed points of the mapping

$$\mathcal{T}[X](t) = X^0 + V^0 t + \int_0^t (t - \sigma) A(\sigma, X(\sigma)) \mathrm{d}\sigma.$$

We stress that here A depends on ψ.

Further, we prove in Section 7 that for $q \geq 7/2$, some $\tau > 0$ and any fixed $X \in \mathcal{B}_{\mathrm{nuc}}(\tau)$, the Cauchy problem (1.4) has a unique solution $\psi \in \mathcal{B}_{\mathrm{el}}(\tau)$, and the mapping $\mathcal{E}[X] = \psi$ is bounded and continuous as a map from $C^0([0, \tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})$ to $C^0([0, \tau]; L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))$. Similarly, solutions are constructed as fixed points of the mapping

$$\mathcal{F}[\psi](t) = U(t, 0)\psi^0 - i \int_0^t U(t, \sigma) V_{\mathrm{Hx}}[\rho]\psi(\sigma) \mathrm{d}\sigma.$$

Using results from Sections 2 and 5 and [31], we show that fixed points of this mapping are strong solutions to (1.4).

We then prove in Section 8 that for $q \geq 7/2$ and some $\tau > 0$, the initial-value problem associated with the problem (1.1) has a solution (ψ, X) in $\mathcal{X}(\tau)$. To this end, we construct the mapping

$$\mathcal{K} : \mathcal{B}_{\mathrm{nuc}}(\tau) \longrightarrow \mathcal{B}_{\mathrm{nuc}}(\tau), \qquad \mathcal{K} = \mathcal{I} \circ \mathcal{N} \circ \mathcal{E},$$

where

$$\mathcal{I} : \mathcal{B}_{\mathrm{nuc}}(\tau) \cap C^2([0, \tau]; \mathbb{R}^{3N_{\mathrm{nuc}}}) \longleftrightarrow \mathcal{B}_{\mathrm{nuc}}(\tau)$$

is the inclusion into $\mathcal{B}_{\mathrm{nuc}}(\tau)$; we then apply a Schauder-type argument to $\mathcal{K}$, in the spirit of [8]. Unlike in [8], we equip $\mathcal{B}_{\mathrm{nuc}}(\tau)$ with a weaker C^0-topology, which takes into account nuclear repulsion. The remainder of this section is devoted to uniqueness.

Finally, the Appendix is devoted to the notation we systematically use, comprising that for the norms on different function spaces, such as $H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$ and Lorentz spaces.

1.4. Related questions

Theorem 1.1 can be generalised to LDA-type nonlinearities which are either sufficiently smooth at the origin $\rho = 0$ or enjoy H^2-Lipschitz estimates such as those obtained in this article. This is the case, for instance, of $\lambda_1 \rho^{q_1 - 1} - \lambda_2 \rho^{q_2 - 1}$ with $q_1, q_2 \geq 7/2$ and $\lambda_1, \lambda_2 > 0$, which share a similar structure with nonlinearities involved in various well-known models in quantum mechanics, such as the Thomas–Fermi–Dirac–von Weizsäcker model [21]. For this particular example, with one working with the same functional setting, it would be interesting to explore, for certain ranges of exponents, the occurrence of either a blow-up at finite time in the norm of the solutions or the existence of maximal solutions defined for all $t \geq 0$; see [11,12].

Also, it would be interesting to identify a functional setting (and a possibly different proof)—the most natural one would certainly be H^1—in order to capture the physically relevant exponent $q = 4/3$, which is

not covered in the present work. We wonder if a suitable regularisation "at the origin" of the LDA term for $q = 4/3$ would allow one to cover this case as a result of a limit process.

2. Preliminaries

The first observation in this section is that the Newton potential

$$G[\phi_1, \phi_2] := (\overline{\phi_1}\,\phi_2) * |\cdot|^{-1} \tag{2.1}$$

solution to

$$-\Delta_x G = 4\pi\overline{\phi_1}\phi_2 \tag{2.2}$$

defines a mapping $H^2 \times H^2 \longrightarrow W^{2,\infty}$.

Lemma 2.1. *For all $i, j \in \{1, 2, 3\}$ and every $x \in \mathbb{R}^3$, it holds that*

$$|G[\phi_1, \phi_2](x)| \lesssim \|\phi_1\|_{L^2}\|\nabla_x\phi_2\|_{L^2}, \tag{2.3}$$

$$|\partial_i G[\phi_1, \phi_2](x)| \lesssim \|\nabla_x\phi_1\|_{L^2}\|\nabla_x\phi_2\|_{L^2}, \tag{2.4}$$

$$|\partial_{ij} G[\phi_1, \phi_2](x)| \lesssim \|\phi_1\|_{H^2}\|\phi_2\|_{H^2}. \tag{2.5}$$

Proof. By Hardy's inequality (A.2) and the properties

$$\partial_i G[\phi_1, \phi_2] = (\overline{\phi_1}\phi_2) * (x_i|x|^{-3}),$$

$$\partial_{ij} G[\phi_1, \phi_2] = [(\partial_i\overline{\phi_1})\phi_2 + \overline{\phi_1}(\partial_j\phi_2)] * (x_i|x|^{-3}),$$

for all i, j and $x \in \mathbb{R}^3$, it holds that

$$|G[\phi_1, \phi_2](x)| = |(\phi_1, |\cdot -x|^{-1}\phi_2)_{L^2}| \lesssim \|\phi_1\|_{L^2}\|\nabla_x\phi_2\|_{L^2},$$

$$|\partial_i G[\phi_1, \phi_2](x)| \leq (|\cdot -x|^{-1}|\phi_1|, |\cdot -x|^{-1}|\phi_2|)_{L^2} \lesssim \|\nabla_x\phi_1\|_{L^2}\|\nabla_x\phi_2\|_{L^2},$$

$$|\partial_{ij} G[\phi_1, \phi_2](x)| \leq (|\cdot -x|^{-1}|\partial_i\phi_1|, |\cdot -x|^{-1}|\phi_2|)_{L^2} + (|\cdot -x|^{-1}|\phi_1|, |\cdot -x|^{-1}|\partial_j\phi_2|)_{L^2}$$

$$\lesssim \|\nabla_x\partial_i\phi_1\|_{L^2}\|\nabla_x\phi_2\|_{L^2} + \|\nabla_x\phi_1\|_{L^2}\|\nabla_x\partial_j\phi_2\|_{L^2}$$

$$\lesssim \|\phi_1\|_{H^2}\|\phi_2\|_{H^2}.$$

This concludes the proof. □

The following lemma generalises Lemma 3 in [8], and provides us with useful bounds on the functions $f_K^{k\ell} : \mathbb{R}^{3N_{\mathrm{nuc}}} \longrightarrow \mathbb{C}^3$ defined as

$$f_K^{k\ell} := \nabla_{X_K}(\psi_k, V_{\mathrm{ext}}[X]\psi_\ell)_{L^2};$$

namely,

$$f_K^{k\ell}(X) = -Z_K\left(\psi_k, \frac{\cdot - X_K}{|\cdot - X_K|^3}\psi_\ell\right)_{L^2}.$$

Note that $f_K^{k\ell}$ effectively depends only on the position X_K of the Kth nucleus, and that

$$A_K^1 = -\frac{1}{M_K}\sum_{k=1}^{N_{\mathrm{el}}} f_K^{kk}.$$

Lemma 2.2. *For all $\psi_k, \psi_\ell \in H^2$, it holds that*

$$\left\| f_K^{k\ell} \right\|_{L^\infty(\mathbb{R}^{3N_{\mathrm{nuc}}};\mathbb{C}^3)} \lesssim \|\nabla_x \psi_k\|_{L^2}\|\nabla_x \psi_\ell\|_{L^2}$$

and

$$\left\| D f_K^{k\ell} \right\|_{L^\infty(\mathbb{R}^{3N_{\mathrm{nuc}}};\mathbb{C}^{3\times 3})} \lesssim \|\psi_k\|_{H^2}\|\psi_\ell\|_{H^2}. \tag{2.6}$$

Here, D is the gradient in $\mathbb{R}^{3N_{\mathrm{nuc}}}$. In addition, we have that $f_K^{k\ell} \in W^{1,\infty} \cap C^1$ for all K.

Proof. By Lemma 2.1, $G[\phi_1, \phi_2] \in W^{2,\infty}$ for all $\phi_1, \phi_2 \in H^2$. Using

$$f_K^{k\ell}(X) = -Z_K \nabla_x G[\psi_k, \psi_\ell](X_K),$$

we get

$$\left\| f_K^{k\ell} \right\|_{L^\infty(\mathbb{R}^{3N_{\mathrm{nuc}}};\mathbb{C}^3)} \lesssim \|\nabla_x \psi_k\|_{L^2}\|\nabla_x \psi_\ell\|_{L^2},$$

$$\left\| D f_K^{k\ell} \right\|_{L^\infty(\mathbb{R}^{3N_{\mathrm{nuc}}};\mathbb{C}^{3\times 3})} \lesssim \max_{X_K \in \mathbb{R}^3} \left\| D^2 G[\psi_k, \psi_\ell](X_K) \right\|_{\mathbb{C}^{3\times 3}} \lesssim \|\psi_k\|_{H^2}\|\psi_\ell\|_{H^2}.$$

This shows that $f_K^{k\ell} \in W^{1,\infty}$. By Sobolev's embedding in Hölder spaces, $\overline{\psi_k}\psi_\ell \in C_{\mathrm{loc}}^{0,\alpha}$. With the use of (2.1) from Lemma 2.1 and standard elliptic regularity, it holds that $G[\psi_k, \psi_\ell] \in C^2$, by which $f_K^{k\ell} \in C^1$. $\square$

In what follows we recall some results on the existence of the propagator for the linear parts of the Kohn–Sham-type Hamiltonian $H[X(t), \rho]$ for $t \in [0, \Theta]$, with $0 < \Theta < \infty$, for a given nuclear configuration $X \in C^1([0, \Theta]; \mathbb{R}^{3N_{\mathrm{nuc}}})$.

For some $X \in C^1([0, \Theta]; \mathbb{R}^{3N_{\mathrm{nuc}}})$ and $0 < \Theta < \infty$ fixed, we consider the family of linear time-dependent Hamiltonians $\{H^{\mathrm{lin}}(t), t \in [0, \Theta]\} \subset \mathcal{L}(H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}); L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))$:

$$H^{\mathrm{lin}}(t) := -\tfrac{1}{2}\Delta_x + V(t), \tag{2.7}$$

where

$$V(t, \cdot) := V_{\mathrm{ext}}[X(t)]. \tag{2.8}$$

Note that $H^{\mathrm{lin}}(t)$ is the linear part of $H[X(t), \rho]$, and that for any fixed t it is a self-adjoint operator on $L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$. We emphasise that these expressions depend on the time evolution of the nuclear configuration X. This family of Hamiltonians is naturally associated with the Cauchy problem

$$i\dot\psi = H^{\mathrm{lin}}(t)\psi, \qquad \psi(s) = \psi^0,$$

on a time interval $[0, \Theta]$ for some $s \in [0, \Theta]$. Equivalently, we can formulate the above as an integral equation

$$\psi(t) = U_0(t-s)\psi^0 - i\int_s^t U_0(t-\sigma)V(\sigma)\psi(\sigma)\mathrm{d}\sigma, \tag{2.9}$$

where

$$U_0(t) := \exp(it\Delta_x/2)$$

is the free propagator (i.e. the propagator for the free particle), which is an evolution operator on $H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$. The following lemma is in the spirit of Lemma 4 in [8], which in turn is based on Corollary 1.2 properties (1), (2) and (4), Theorem 1.1 property (2) and Theorem 1.3 properties (5) and (6) in [31].

Lemma 2.3. *For the family of Hamiltonians* $\{H^{\mathrm{lin}}(t), t \in [0, \Theta]\}$, *there exists a unique family of linear evolution operators*

$$U(t, s) : L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}) \longrightarrow L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}), \qquad t, s \in [0, \Theta],$$

such that

$$\psi(t) := U(t, s)\psi^0$$

solves (2.9) *on* $[0, \Theta]$ *for all* $\psi^0 \in H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$, *with*

$$\|\psi(t)\|_{L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})} = \|\psi^0\|_{L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})},$$

for all $t \in [0, \Theta]$. *Moreover, this family has the following properties:*

   (i) $U(t, s)U(s, r) = U(t, r)$ *for all* $t, s, r \in [0, \Theta]$.
  (ii) $U(t, t) = \mathrm{Id}$ *for all* $t \in [0, \Theta]$.
 (iii) $U(t, s)$ *is a unitary operator on* $L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$ *for all* $t, s \in [0, \Theta]$:

$$\|U(t, s)\psi\|_{L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})} = \|\psi\|_{L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})}.$$

  (iv) *For all* $f \in L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$, $((t, s) \longmapsto U(t, s)f) : [0, \Theta]^2 \longrightarrow L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$ *is a continuous mapping.*
   (v) $U(t, s) \in \mathcal{L}(H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))$ *for all* $(t, s) \in [0, \Theta]^2$.
  (vi) *For all* $f \in H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$, $((t, s) \longmapsto U(t, s)f) : [0, \Theta]^2 \longrightarrow H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$ *is a continuous mapping.*
 (vii) *For all* $f \in H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$, *the mapping* $(t, s) \longmapsto U(t, s)f$ *is an element in* $C^1([0, \Theta]^2; L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))$, *and the following equations hold in* $L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$:

$$i\frac{\partial}{\partial t}(U(t, s)f) = H^{\mathrm{lin}}(t)U(t, s)f,$$

$$i\frac{\partial}{\partial s}(U(t, s)f) = -U(t, s)H^{\mathrm{lin}}(s)f.$$

(viii) *For all* $\gamma > 0$, *there is a constant* $B_{\Theta, \gamma}$ *of the form*

$$B_{\Theta, \gamma} = A_\gamma^{1 + C_\gamma \Theta}, \qquad A_\gamma, C_\gamma > 0,$$

such that if

$$\|\dot{X}\|_{C^0([0, \Theta]; \mathbb{R}^{3N_{\mathrm{nuc}}})} \leq \gamma,$$

then for all $t, s \in [0, \Theta]$,

$$\|U(t, s)\|_{\mathcal{L}(H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))} \leq B_{\Theta, \gamma}.$$

Proof. The result in the case $N_{\text{el}} = N_{\text{nuc}} = 1$ was proved in [8, Lemma 4]. We observe that the argument in [8] is robust enough to be easily adapted to our more general context of arbitrary $N_{\text{el}}, N_{\text{nuc}} \in \mathbb{N}$. Indeed, since the linear Hamiltonians $H^{\text{lin}}(t)$ do not depend on an electronic configuration ψ, and act on every element ψ_k independently, the result for general N_{el} follows from the case $N_{\text{el}} = 1$. In particular, properties (i)–(vii) can be justified with an obvious adaptation of the case $N_{\text{el}} = 1$ proved in [31, Corollary 1.2 (1), (2) and (4), Theorem 1.1 (2) and Theorem 1.3 (5) and (6)]. Property (viii) can be justified also by one arguing exactly as for the case $N_{\text{el}} = N_{\text{nuc}} = 1$ in [8, Lemma 4], observing that our additional terms in the expression for V can be estimated in the same way. $\square$

3. Definition of the electronic feasible region $\mathcal{B}_{\text{el}}$

Let $\tau > 0$ be finite and define

$$\gamma := |V^0| + 1, \tag{3.1}$$

where the term "+1" allows us to cover the case $V^0 = 0$. Let us consider $B_{\tau,\gamma}$ as given in Lemma 2.3 with $\Theta = \tau$, and where γ is as above. We can therefore define the radius

$$\alpha(\tau) := 2B_{\tau,\gamma} \|\psi^0\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\text{el}}})}$$

for the ball centred at the initial configuration $\psi^0 \in H^2(\mathbb{R}^3;\mathbb{C}^{N_{\text{el}}})$:

$$B_\alpha(\psi^0) = \{\psi \in H^2(\mathbb{R}^3;\mathbb{C}^{N_{\text{el}}}) | \|\psi - \psi^0\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\text{el}}})} \leq \alpha\}.$$

Finally, let us define the electronic feasible region for the time interval $[0,\tau]$ as

$$\mathcal{B}_{\text{el}}(\tau) := \{\psi \in C^1([0,\tau];L^2(\mathbb{R}^3;\mathbb{C}^{N_{\text{el}}})) \cap C^0([0,\tau];B_\alpha(\psi^0)) \mid \psi(0) = \psi^0\},$$

equipped with the $C^0([0,\tau];L^2(\mathbb{R}^3;\mathbb{C}^{N_{\text{el}}}))$ norm, which is designed to contain solutions ψ to the Cauchy problem associated with (1.4) with $\psi(0) = \psi^0$ on the time interval $[0,\tau]$, which we call "feasible electronic configurations".

4. Definition of the nuclear feasible region $\mathcal{B}_{\text{nuc}}$

For all $0 < \varepsilon < \min_{K \neq L}\{|X_K^0 - X_L^0|\}$, we set

$$\delta(\tau) := \frac{\min_{K \neq L}\{|X_K^0 - X_L^0|\} - \min\{\delta_{\text{rep}}(\tau),\varepsilon\}}{2} > 0,$$

where

$$\delta_{\text{rep}}(\tau) := \left[\left(\sum_{K=1}^{N_{\text{nuc}}} M_K |V_K^0|^2 + \sum_{K,L=1,L\neq K}^{N_{\text{nuc}}} \frac{Z_K Z_L}{|X_K^0 - X_L^0|}\right)e^\tau + 16\sum_{K=1}^{N_{\text{nuc}}} \frac{Z_K^2}{M_K}\|\psi\|^2_{C^0([0,\tau];H^1(\mathbb{R}^3;\mathbb{C}^{N_{\text{el}}}))}(e^\tau - 1)\right]^{-1}$$

arises from a repulsion argument, given in Lemma 4.1. Note that $\varepsilon > 0$ ensures the strict positivity of $\delta(\tau)$, which defines the radius for the ball centred at the initial configuration $X^0 \in \mathbb{R}^{3N_{\text{nuc}}}$, with $X_K^0 \neq X_L^0$ for $K \neq L$:

$$B_\delta(X^0) = \{X \in \mathbb{R}^{3N_{\text{nuc}}} | |X - X^0| \leq \delta\}.$$

Then, by the triangle inequality, for all $X \in B_\delta(X^0)$ and $K \neq L$, it holds that

$$|X_K - X_L| \geq \min_{K' \neq L'} |X_{K'}^0 - X_{L'}^0| - 2|X - X^0| \geq \min_{K' \neq L'} |X_{K'}^0 - X_{L'}^0| - 2\delta(\tau) = \min\{\delta_{\text{rep}}(\tau), \varepsilon\} > 0.$$

We define the nuclear feasible region for the time interval $[0, \tau]$ as

$$\mathcal{B}_{\text{nuc}}(\tau) := \{X \in C^1([0, \tau]; B_\delta(X^0)) \,|\, X(0) = X^0, \dot{X}(0) = V^0, \|\dot{X}\|_{C^0([0,\tau];\mathbb{R}^{3N_{\text{nuc}}})} \leq \gamma\},$$

with γ as in (3.1). This region is equipped with the $C^0([0, \tau]; \mathbb{R}^{3N_{\text{nuc}}})$ topology, and is designed to contain short-time solutions X to the Cauchy problem associated with (1.6) with $X(0) = X^0$ and $\dot{X}(0) = V^0$ on the interval $[0, \tau]$, which we call "feasible nuclear configurations".

This definition of $\delta(\tau)$ is suggested by an a priori lower bound on the nuclear distances $|X_K(t) - X_L(t)|$, $K \neq L$, which is based on Grönwall's lemma. We have the following lemma.

Lemma 4.1. *Fix $\psi \in C^0([0, \tau]; H^1(\mathbb{R}^3; \mathbb{C}^{N_{\text{el}}}))$, and $X^0 \in \mathbb{R}^{3N_{\text{nuc}}}$ such that $X_K^0 \neq X_L^0$ for $K \neq L$. Let X solve (1.6), and let $X(0) = X^0$. Then, for all $t \in [0, \tau]$ and $K \neq L$,*

$$|X_K(t) - X_L(t)| \geq \delta_{\text{rep}}(\tau).$$

Proof. Writing the momenta $P_K := M_K \dot{X}_K$, we define the classical reduced Hamiltonian

$$\mathcal{H}_{\text{nn}}(X, P) := \frac{1}{2} \sum_{K=1}^{N_{\text{nuc}}} \frac{|P_K|^2}{M_K} + W_{\text{nn}}(X),$$

with W_{nn} as in (1.7). Fix $\psi \in C^0([0, \tau]; H^1(\mathbb{R}^3; \mathbb{C}^{N_{\text{el}}}))$. Now

$$\frac{\mathrm{d}}{\mathrm{d}t}[\mathcal{H}_{\text{nn}}(X, P)] = \sum_{K=1}^{N_{\text{nuc}}} [\nabla_{X_K} \mathcal{H}_{\text{nn}}(X, P) \cdot \dot{X}_K + \nabla_{P_K} \mathcal{H}_{\text{nn}}(X, P) \cdot \dot{P}_K]$$

$$= \sum_{K=1}^{N_{\text{nuc}}} \frac{P_K}{M_K} \cdot \{\nabla_{X_K}[W_{\text{nn}}(X)] + M_K \ddot{X}_K\}$$

$$\overset{(1.6)}{=} -\sum_{K=1}^{N_{\text{nuc}}} \frac{P_K}{M_K} \cdot (\nabla_{X_K} V_{\text{ext}}[X], \rho)_{L^2}$$

$$\leq \sum_{K=1}^{N_{\text{nuc}}} \frac{1}{2M_K} [|P_K|^2 + |(\nabla_{X_K} V_{\text{ext}}[X], \rho)_{L^2}|^2]$$

$$\leq \mathcal{H}_{\text{nn}}(X, P) + 8 \sum_{K=1}^{N_{\text{nuc}}} \frac{Z_K^2}{M_K} \|\psi\|_{C^0([0,\tau];H^1(\mathbb{R}^3;\mathbb{C}^{N_{\text{el}}}))}^2, \tag{4.1}$$

by which, with the use of Grönwall's inequality, on $[0, \tau]$

$$\mathcal{H}_{\text{nn}}(X, P) \leq \frac{1}{2} e^\tau \left[\sum_{K=1}^{N_{\text{nuc}}} M_K |V_K^0|^2 + \sum_{K,L=1,L\neq K}^{N_{\text{nuc}}} \frac{Z_K Z_L}{|X_K^0 - X_L^0|} + 16 \sum_{K=1}^{N_{\text{nuc}}} \frac{Z_K^2}{M_K} \|\psi\|_{C^0([0,\tau];H^1(\mathbb{R}^3;\mathbb{C}^{N_{\text{el}}}))}^2 \right]$$

$$- 8 \sum_{K=1}^{N_{\text{nuc}}} \frac{Z_K^2}{M_K} \|\psi\|_{C^0([0,\tau];H^1(\mathbb{R}^3;\mathbb{C}^{N_{\text{el}}}))}^2. \tag{4.2}$$

In (4.1), we use the fact that, by Hardy's inequality, for all $K = 1, \ldots, N_{\mathrm{nuc}}$,

$$
|(\nabla_{X_K} V_{\mathrm{ext}}[X], \rho)_{L^2}| = \left| \left(-Z_K \frac{\cdot - X_K}{|\cdot - X_K|^3}, \rho \right)_{L^2} \right| \leq Z_K \sum_{k=1}^{N_{\mathrm{el}}} \left\| |\cdot - X_K|^{-1} \psi_k \right\|_{L^2}
$$

$$
\leq 2Z_K \sum_{k=1}^{N_{\mathrm{el}}} \|\nabla_x \psi_k\|_{L^2} \leq 2\sqrt{2} Z_K \|\nabla_x \psi\|_{L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})} \leq 2\sqrt{2} Z_K \|\psi\|_{H^1(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})}.
$$

Estimate (4.2) is enough to conclude the proof, as for all $K \neq L$ we have

$$
\tfrac{1}{2} \frac{1}{|X_K - X_L|} \leq \frac{1}{Z_K Z_L} W_{\mathrm{nn}}(X) \leq W_{\mathrm{nn}}(X) \leq \mathcal{H}_{\mathrm{nn}}(X, P). \quad \square
$$

Remark 4.1. A similar argument yields an a priori estimate of the nuclear velocity $\dot{X}$.

5. Lipschitz estimates

In this section, we obtain Lipschitz estimates on the mapping $\psi \longmapsto V_{\mathrm{Hx}}[\rho]\psi := (V_{\mathrm{H}}[\rho] + V_{\mathrm{x}}[\rho])\psi$.

Lemma 5.1 (*Lipschitz estimates on the Hartree term*). *For all* $\psi, \psi' \in H^1(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$, *with* $\rho' := |\psi'|^2$,

$$
\|V_{\mathrm{H}}[\rho]\psi - V_{\mathrm{H}}[\rho']\psi'\|_{L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})} \lesssim \sqrt{N_{\mathrm{el}}} \|\psi - \psi'\|_{L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})} \times
$$

$$
\times \left[\sum_{k=1}^{N_{\mathrm{el}}} (\|\nabla_x \psi_k\|_{L^2(\mathbb{R}^3; \mathbb{C}^3)} + \|\nabla_x \psi'_k\|_{L^2(\mathbb{R}^3; \mathbb{C}^3)}) \|\psi'\|_{L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})} + \sum_{\ell=1}^{N_{\mathrm{el}}} \|\psi_\ell\|_{L^2} \|\nabla_x \psi_\ell\|_{L^2(\mathbb{R}^3; \mathbb{C}^3)} \right]. \quad (5.1)
$$

Moreover, for all $\psi, \psi' \in H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$,

$$
\|V_{\mathrm{H}}[\rho]\psi\|_{H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})} \lesssim \sqrt{N_{\mathrm{el}}} \sum_{k=1}^{N_{\mathrm{el}}} \|\psi_k\|_{H^1}^2 \|\psi\|_{H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})}, \quad (5.2)
$$

$$
\|V_{\mathrm{H}}[\rho]\psi - V_{\mathrm{H}}[\rho']\psi'\|_{H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})} \lesssim \sqrt{N_{\mathrm{el}}} \|\psi - \psi'\|_{H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})} \times
$$

$$
\times \sum_{k=1}^{N_{\mathrm{el}}} [(\|\psi_k\|_{H^1} + \|\psi'_k\|_{H^1}) \|\psi'\|_{H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})} + \|\psi_k\|_{H^1}^2]. \quad (5.3)
$$

Proof.

Proof of (5.1).

By adding and subtracting the term $(|\psi'_\ell|^2 * |\cdot|^{-1})\psi'_k$, we can write for all $k = 1, \ldots, N_{\mathrm{el}}$,

$$
\|(V_{\mathrm{H}}[\rho]\psi - V_{\mathrm{H}}[\rho']\psi')_k\|_{L^2} \leq \sum_{\ell=1}^{N_{\mathrm{el}}} [\underbrace{\|(|\psi_\ell|^2 * |\cdot|^{-1})(\psi_k - \psi'_k)\|_{L^2}}_{=: \, (\mathrm{I})} + \underbrace{\|((|\psi_\ell|^2 - |\psi'_\ell|^2) * |\cdot|^{-1})\psi'_k\|_{L^2}}_{=: \, (\mathrm{II})}]. \quad (5.4)
$$

With the use of the Cauchy–Schwarz inequality in (5.5) and (5.7), Hardy's inequality in (5.6) and (5.8), and the triangle inequality in (5.8),

$$
(\mathrm{I}) \leq \||\psi_\ell|^2 * |\cdot|^{-1}\|_{L^\infty} \|\psi_k - \psi'_k\|_{L^2} \leq \operatorname*{esssup}_{x \in \mathbb{R}^3} \{|(|\psi_\ell|, |\cdot - x|^{-1}|\psi_\ell|)_{L^2}|\} \|\psi - \psi'\|_{L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})}
$$

$$\leq \operatorname*{esssup}_{x\in\mathbb{R}^3}\{\|\psi_\ell\|_{L^2}\||\cdot-x|^{-1}\psi_\ell\|_{L^2}\}\|\psi-\psi'\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \tag{5.5}$$

$$\lesssim \|\psi_\ell\|_{L^2}\|\nabla_x\psi_\ell\|_{L^2(\mathbb{R}^3;\mathbb{C}^3)}\|\psi-\psi'\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \tag{5.6}$$

and

$$\begin{aligned}
(\mathrm{II}) &\leq \|(|\psi_\ell|^2-|\psi'_\ell|^2)*|\cdot|^{-1}\|_{L^\infty}\|\psi'_k\|_{L^2}\\
&\leq \operatorname*{esssup}_{x\in\mathbb{R}^3}\{|(|\psi_\ell|-|\psi'_\ell|,|\cdot-x|^{-1}(|\psi_\ell|+|\psi'_\ell|))_{L^2}|\}\|\psi'\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}\\
&\leq \operatorname*{esssup}_{x\in\mathbb{R}^3}\{\||\psi_\ell|-|\psi'_\ell|\|_{L^2}(\||\cdot-x|^{-1}\psi_\ell\|_{L^2}+\||\cdot-x|^{-1}\psi'_\ell\|_{L^2})\}\|\psi'\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \tag{5.7}\\
&\lesssim (\|\nabla_x\psi_\ell\|_{L^2(\mathbb{R}^3;\mathbb{C}^3)}+\|\nabla_x\psi'_\ell\|_{L^2(\mathbb{R}^3;\mathbb{C}^3)})\|\psi'\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}\|\psi_\ell-\psi'_\ell\|_{L^2} \tag{5.8}\\
&\lesssim (\|\nabla_x\psi_\ell\|_{L^2(\mathbb{R}^3;\mathbb{C}^3)}+\|\nabla_x\psi'_\ell\|_{L^2(\mathbb{R}^3;\mathbb{C}^3)})\|\psi'\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}\|\psi-\psi'\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}. \tag{5.9}
\end{aligned}$$

Combining (5.6) and (5.9) with (5.4), we get (5.1).

Proof of (5.2).

With G as in (2.1), we can write

$$V_{\mathrm{H}}[\rho]\psi = \sum_{k=1}^{N_{\mathrm{el}}} G[\psi_k,\psi_k]\psi.$$

Note that for all $\phi_1,\phi_2,\phi_3\in H^2$,

$$\Delta_x(G[\phi_1,\phi_2]\phi_3) \stackrel{(2.2)}{=} G[\phi_1,\phi_2]\Delta_x\phi_3 + 2\nabla_x(G[\phi_1,\phi_2])\cdot\nabla_x\phi_3 - 4\pi\overline{\phi_1}\phi_2\phi_3.$$

Using (2.3) and (2.4) from Lemma 2.1, we obtain

$$\|G[\phi_1,\phi_2]\phi_3\|_{L^2} \leq \|G[\phi_1,\phi_2]\|_{L^\infty}\|\phi_3\|_{L^2} \lesssim \|\phi_1\|_{H^1}\|\phi_2\|_{H^1}\|\phi_3\|_{H^1},$$

$$\|G[\phi_1,\phi_2]\Delta_x\phi_3\|_{L^2} \leq \|G[\phi_1,\phi_2]\|_{L^\infty}\|\Delta_x\phi_3\|_{L^2} \lesssim \|\phi_1\|_{H^1}\|\phi_2\|_{H^1}\|\phi_3\|_{H^2},$$

$$\|\nabla_x(G[\phi_1,\phi_2])\cdot\nabla_x\phi_3\|_{L^2} \leq \|G[\phi_1,\phi_2]\|_{W^{1,\infty}}\|\nabla_x\phi_3\|_{L^2(\mathbb{R}^3;\mathbb{C}^3)} \lesssim \|\phi_1\|_{H^1}\|\phi_2\|_{H^1}\|\phi_3\|_{H^1}.$$

On the other hand, by Hölder's and Sobolev's inequalities,

$$\|\overline{\phi_1}\phi_2\phi_3\|_{L^2} \leq \|\phi_1\|_{L^6}\|\phi_2\|_{L^6}\|\phi_3\|_{L^6} \lesssim \|\phi_1\|_{H^1}\|\phi_2\|_{H^1}\|\phi_3\|_{H^1}.$$

This gives for all $k=1,\ldots,N_{\mathrm{el}}$,

$$\|(V_{\mathrm{H}}[\rho]\psi)_k\|_{H^2} \leq \sum_{\ell=1}^{N_{\mathrm{el}}}\|G[\psi_\ell,\psi_\ell]\psi_k\|_{H^2} \lesssim \sum_{\ell=1}^{N_{\mathrm{el}}}\|\psi_\ell\|_{H^1}^2\|\psi\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}.$$

By our combining these estimates, (5.2) follows.

Proof of (5.3).

As in the proof of (5.1), we bound for all $k=1,\ldots,N_{\mathrm{el}}$,

$$\|\Delta_x(V_{\mathrm{H}}[\rho]\psi - V_{\mathrm{H}}[\rho']\psi')_k\|_{L^2} \leq \sum_{\ell=1}^{N_{\mathrm{el}}}\{\underbrace{\|\Delta_x[G[\psi_\ell,\psi_\ell](\psi_k-\psi'_k)]\|_{L^2}}_{=:\,(\mathrm{I})} + \underbrace{\|\Delta_x[G[|\psi_\ell|+|\psi'_\ell|,|\psi_\ell|-|\psi'_\ell|]\psi'_k]\|_{L^2}}_{=:\,(\mathrm{II})}\}.$$

As for (5.2), we can bound (I) using $(\phi_1, \phi_2, \phi_3) = (\psi_\ell, \psi_\ell, \psi_k - \psi_k')$ and can bound (II) using $(\phi_1, \phi_2, \phi_3) = (|\psi_\ell| + |\psi_\ell'|, |\psi_\ell| - |\psi_\ell'|, \psi_k)$. Hence, by the triangle inequality,

$$\|\Delta_x(V_{\mathrm{H}}[\rho]\psi - V_{\mathrm{H}}[\rho']\psi')\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \lesssim (\mathrm{B}) :=$$

$$= \sqrt{N_{\mathrm{el}}} \sum_{k=1}^{N_{\mathrm{el}}} \big[\|\psi_k\|_{H^1}^2 + (\|\psi_k\|_{H^1} + \|\psi_k'\|_{H^1}) \|\psi'\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \big] \|\psi - \psi'\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}. \tag{5.10}$$

On the other hand, by (5.1) we also have

$$\|V_{\mathrm{H}}[\rho]\psi - V_{\mathrm{H}}[\rho']\psi'\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \lesssim (\mathrm{B}). \tag{5.11}$$

Hence, by (5.10) and (5.11), (5.3) immediately follows. □

By the Cauchy–Schwarz inequality, for all $\mathbb{C}^{N_{\mathrm{el}}}$-valued functions ψ, ψ', we have

$$|\psi \cdot \nabla_x \psi'| \le |\psi| |\nabla_x \psi'|. \tag{5.12}$$

Lemma 5.2 *(Mean-value estimates for the density).* *For all $a \ge 1/2$, we have*

$$|\rho^a - \rho'^{\,a}| \lesssim_a (\|\rho\|_{L^\infty}^{a-1/2} + \|\rho'\|_{L^\infty}^{a-1/2}) |\psi - \psi'|. \tag{5.13}$$

Proof. By the fundamental theorem of calculus

$$|\rho^a - \rho'^{\,a}| = ||\psi|^{2a} - |\psi'|^{2a}| = \left| \int_0^1 \frac{\mathrm{d}}{\mathrm{d}t} [|\psi' + t(\psi - \psi')|^{2a}] \mathrm{d}t \right|$$

$$\lesssim_a (|\psi| + |\psi'|)^{2a-1} |\psi - \psi'| \lesssim_a (|\psi|^{2a-1} + |\psi'|^{2a-1}) |\psi - \psi'|$$

$$= (\rho^{a-1/2} + \rho'^{\,a-1/2}) |\psi - \psi'|,$$

which yields (5.13). □

Lemma 5.3 *(Mean-value estimates for the density gradient).* *For all $b \ge 3/2$, it holds that*

$$|\nabla_x(\rho^b) - \nabla_x(\rho'^{\,b})| \lesssim_b (Q_1 |\nabla_x \psi| + Q_2 |\nabla_x \psi'|) |\psi - \psi'| + Q_3 |\nabla_x \psi - \nabla_x \psi'|, \tag{5.14}$$

where

$$Q_1 = \rho^{b-1}, \qquad Q_2 = (\rho^{b-3/2} + \rho'^{\,b-3/2}) \rho'^{\,1/2}, \qquad Q_3 = \rho^{b-1} \rho'^{\,1/2}.$$

Proof. Using $\nabla_x \rho = \nabla_x \psi \cdot \overline{\psi} + \psi \cdot \nabla_x(\overline{\psi})$ and (5.12) for the pair $(\psi, \overline{\psi})$, we have

$$|\nabla_x \rho| \lesssim \rho^{1/2} |\nabla_x \psi|. \tag{5.15}$$

Since

$$|\nabla_x(\rho^b)| \lesssim_b \rho^{b-1} |\nabla_x \rho|,$$

adding and subtracting the term $\rho^{b-1} \nabla_x \rho'$, we get for all $b \ge 1$,

$$|\nabla_x(\rho^b) - \nabla_x(\rho'^{\,b})| \lesssim_b \rho^{b-1}|\nabla_x\rho - \nabla_x\rho'| + |\rho^{b-1} - \rho'^{\,b-1}||\nabla_x\rho'|.$$

By adding and subtracting $\psi' \cdot \nabla_x(\overline{\psi})$ and $\overline{\psi'} \cdot \nabla_x\psi$, and using (5.12) for the pairs $(\psi - \psi', \overline{\psi})$, $(\psi', \overline{\psi - \psi'})$, $(\overline{\psi - \psi'}, \psi)$ and $(\overline{\psi'}, \psi - \psi')$, we get

$$
\begin{aligned}
|\nabla_x\rho - \nabla_x\rho'| &= |\psi \cdot \nabla_x(\overline{\psi}) - \psi' \cdot \nabla_x(\overline{\psi'}) + \overline{\psi} \cdot \nabla_x\psi - \overline{\psi'} \cdot \nabla_x(\psi')| \\
&\leq |(\psi - \psi') \cdot \nabla_x(\overline{\psi})| + |\psi' \cdot \nabla_x(\overline{\psi - \psi'})| + |(\overline{\psi - \psi'}) \cdot \nabla_x\psi| + |\overline{\psi'} \cdot \nabla_x(\psi - \psi')| \\
&\lesssim |\nabla_x\psi||\psi - \psi'| + \rho'^{\,1/2}|\nabla_x\psi - \nabla_x\psi'|.
\end{aligned}
\tag{5.16}
$$

By (5.13) with $a = b - 1 \geq 1/2$ and (5.15) for ρ', we get

$$|\rho^{b-1} - \rho'^{\,b-1}||\nabla_x\rho'| \lesssim_b (\rho^{b-3/2} + \rho'^{\,b-3/2})\rho'^{\,1/2}|\nabla_x\psi'||\psi - \psi'|.$$

Putting these estimates together, we get (5.14). $\quad\square$

Lemma 5.4 *(Lipschitz estimates on the local nonlinearity). Let $q \in [1, \infty)$ and $\lambda \in \mathbb{R}$. For any fixed $p \in [1, \infty]$ and for all $\psi, \psi' \in H^2(\mathbb{R}^3; \mathbb{C}^{N_{el}}) \cap L^p(\mathbb{R}^3; \mathbb{C}^{N_{el}})$, it holds that*

$$\|V_x[\rho]\psi - V_x[\rho']\psi'\|_{L^p(\mathbb{R}^3;\mathbb{C}^{N_{el}})} \lesssim_{q,\lambda} \sum_{k=1}^{N_{el}} \left[\|\psi_k\|_{H^2}^{2(q-1)} + \|\psi_k'\|_{H^2}^{2(q-1)} \right] \|\psi - \psi'\|_{L^p(\mathbb{R}^3;\mathbb{C}^{N_{el}})}. \tag{5.17}$$

Moreover, for all $q \geq 7/2$ and any $\lambda \in \mathbb{R}$, we have

$$\|V_x[\rho]\psi - V_x[\rho']\psi'\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{el}})} \leq \mathcal{L}_{q,\lambda}(\max\{\|\psi\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{el}})}, \|\psi'\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{el}})}\})\|\psi - \psi'\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{el}})}, \tag{5.18}$$

where $\mathcal{L}_{q,\lambda} : \mathbb{R}_0^+ \longrightarrow \mathbb{R}_0^+$ is a polynomial function which vanishes at the origin.

Proof.

Proof of (5.17).

By the fundamental theorem of calculus,

$$
\begin{aligned}
|V_x[\rho]\psi - V_x[\rho']\psi'| &= |\lambda|||\psi|^{2(q-1)}\psi - |\psi'|^{2(q-1)}\psi'| \\
&\lesssim_\lambda \left| \int\limits_0^1 \frac{\mathrm{d}}{\mathrm{d}t}\left[|\psi' + t(\psi - \psi')|^{2(q-1)}(\psi' + t(\psi - \psi'))\right]\mathrm{d}t \right| \\
&\lesssim_q |\psi - \psi'| \int\limits_0^1 |\psi' + t(\psi - \psi')|^{2(q-1)}\mathrm{d}t \\
&\leq (|\psi| + |\psi'|)^{2(q-1)}|\psi - \psi'| \lesssim_q (\rho^{q-1} + \rho'^{\,q-1})|\psi - \psi'|.
\end{aligned}
$$

Since H^2 is embedded into L^∞,

$$\|\rho\|_{L^\infty}^a \lesssim_a \sum_{k=1}^{N_{el}} \|\psi_k\|_{H^2}^{2a} \tag{5.19}$$

for all $a > 0$. By our taking $a = q - 1 > 0$, (5.17) follows.

Proof of (5.18).

Taking $p = 2$ in (5.17), we need only the $L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$ norm of $\Delta_x(V_{\mathrm{x}}[\rho]\psi - V_{\mathrm{x}}[\rho']\psi')$ in addition to get the $H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$ norm estimate. Using the product rule for the Laplacian in $\mathbb{R}^3$, we get

$$\Delta_x(V_{\mathrm{x}}[\rho]\psi - V_{\mathrm{x}}[\rho']\psi') = \lambda\Big\{ \underbrace{\rho^{q-1}\Delta_x\psi - \rho'^{\,q-1}\Delta_x\psi'}_{=: \, (\mathrm{I})} + \underbrace{2[\nabla_x(\rho^{q-1})\cdot\nabla_x\psi - \nabla_x(\rho'^{\,q-1})\cdot\nabla_x\psi']}_{=: \, (\mathrm{II})}$$
$$+ \underbrace{\Delta_x(\rho^{q-1})\psi - \Delta_x(\rho'^{\,q-1})\psi'}_{=: \, (\mathrm{III})} \Big\}, \tag{5.20}$$

which is in $\mathbb{C}^{N_{\mathrm{el}}}$. We discuss the terms one by one.

Term (I).

By adding and subtracting the term $\rho^{q-1}\Delta_x\psi'$ and using (5.13) with $a = q - 1 > 1$, we get

$$|(\mathrm{I})| \le |\rho^{q-1}||\Delta_x\psi - \Delta_x\psi'| + |\rho^{q-1} - \rho'^{\,q-1}||\Delta_x\psi'|$$
$$\lesssim_q A_1|\Delta_x\psi'||\psi - \psi'| + A_2|\Delta_x\psi - \Delta_x\psi'|, \tag{5.21}$$

where

$$A_1 = \|\rho\|_{L^\infty}^{q-3/2} + \|\rho'\|_{L^\infty}^{q-3/2}, \qquad A_2 = \|\rho\|_{L^\infty}^{q-1}.$$

Term (II).

By adding and subtracting the term $\nabla_x(\rho^{q-1})\cdot\nabla_x\psi'$, we get

$$|\nabla_x(\rho^{q-1})\cdot\nabla_x\psi - \nabla_x(\rho'^{\,q-1})\cdot\nabla_x\psi'| \le |\nabla_x(\rho^{q-1})||\nabla_x\psi - \nabla_x\psi'| + |\nabla_x\psi'||\nabla_x(\rho^{q-1}) - \nabla_x(\rho'^{\,q-1})|.$$

We then get

$$|\nabla_x(\rho^{q-1})| = (q-1)|\rho^{q-2}||\nabla_x\rho| \overset{(5.15)}{\lesssim} (q-1)\|\rho\|_{L^\infty}^{q-3/2}|\nabla_x\psi|.$$

Using this and (5.14) with $b = q - 1 > 2$, we get

$$|(\mathrm{II})| \lesssim_q (B_1|\nabla_x\psi||\nabla_x\psi'| + B_2|\nabla_x\psi'|^2)|\psi - \psi'| + (B_3|\nabla_x\psi| + B_4|\nabla_x\psi'|)|\nabla_x\psi - \nabla_x\psi'|, \tag{5.22}$$

where

$$B_1 = \|\rho\|_{L^\infty}^{q-2}, \qquad B_2 = \|\rho'\|_{L^\infty}^{1/2}(\|\rho\|_{L^\infty}^{q-5/2} + \|\rho'\|_{L^\infty}^{q-5/2}),$$
$$B_3 = \|\rho\|_{L^\infty}^{q-3/2}, \qquad B_4 = \|\rho\|_{L^\infty}^{q-2}\|\rho'\|_{L^\infty}^{1/2}.$$

Term (III).

By adding and subtracting the term $\Delta_x(\rho^{q-1})\psi'$, we get

$$|(\mathrm{III})| \le \underbrace{|\Delta_x(\rho^{q-1})|}_{=: \, (\mathrm{a})} |\psi - \psi'| + \underbrace{|\Delta_x(\rho^{q-1}) - \Delta_x(\rho'^{\,q-1})|}_{=: \, (\mathrm{b})} \|\rho'\|_{L^\infty}^{1/2}.$$

Using $\Delta_x\rho = \overline{\psi}\cdot\Delta_x\psi + \overline{\Delta_x\psi}\cdot\psi + 2|\nabla_x\psi|^2$ and the Cauchy–Schwarz inequality, we get

$$|\Delta_x\rho| \lesssim \|\rho\|_{L^\infty}^{1/2}|\Delta_x\psi| + |\nabla_x\psi|^2. \tag{5.23}$$

Hence,

$$(a) \lesssim_q (q-2)|\rho|^{q-3}|\nabla_x\rho|^2 + |\rho|^{q-2}|\Delta_x\rho| \overset{(5.15)}{\lesssim} \|\rho\|_{L^\infty}^{q-2}\big[(2q-3)|\nabla_x\psi|^2 + \|\rho\|_{L^\infty}^{1/2}|\Delta_x\psi|\big].$$

By similar reasoning, we get, by adding and subtracting the terms $\rho^{q-3}|\nabla_x\rho'|^2$ and $\rho^{q-2}\Delta_x\rho'$,

$$(b) \lesssim_q (q-2)\big(|\rho|^{q-3}\underbrace{\big||\nabla_x\rho|^2 - |\nabla_x\rho'|^2\big|}_{=:\,(i)} + \underbrace{|\rho^{q-3} - {\rho'}^{q-3}||\nabla_x\rho'|^2}_{=:\,(ii)}\big) + |\rho|^{q-2}\underbrace{|\Delta_x\rho - \Delta_x\rho'|}_{=:\,(iii)}$$

$$+ \underbrace{|\rho^{q-2} - {\rho'}^{q-2}||\Delta_x\rho'|}_{=:\,(iv)}.$$

By (5.15), we get

$$(i) \le (|\nabla_x\rho| + |\nabla_x\rho'|)|\nabla_x\rho - \nabla_x\rho'|$$

$$\overset{(5.16)}{\lesssim} (\|\rho\|_{L^\infty}^{1/2}|\nabla_x\psi| + \|\rho'\|_{L^\infty}^{1/2}|\nabla_x\psi'|)(|\psi - \psi'||\nabla_x\psi| + \|\rho'\|_{L^\infty}^{1/2}|\nabla_x\psi - \nabla_x\psi'|).$$

Furthermore, using (5.13) with $a = q - 3 \ge 1/2$ (here is where we use the restriction $q \ge 7/2$) and (5.15) for ρ', we get

$$(ii) \lesssim_q (\|\rho\|_{L^\infty}^{q-7/2} + \|\rho'\|_{L^\infty}^{q-7/2})\|\rho'\|_{L^\infty}|\nabla_x\psi'|^2|\psi - \psi'|.$$

In addition, by adding and subtracting the terms $\overline{\psi'} \cdot \Delta_x\psi$ and $\overline{\Delta_x\psi} \cdot \psi'$, and using the triangle and Cauchy–Schwarz inequalities, we get

$$(iii) = \big|2(|\nabla_x\psi|^2 - |\nabla_x\psi'|^2) + (\overline{\psi} - \overline{\psi'}) \cdot \Delta_x\psi + (\Delta_x\psi - \Delta_x\psi') \cdot \overline{\psi'}$$

$$+ (\psi - \psi') \cdot \overline{\Delta_x\psi} + (\overline{\Delta_x\psi} - \overline{\Delta_x\psi'}) \cdot \psi'\big|$$

$$\lesssim (|\nabla_x\psi| + |\nabla_x\psi'|)|\nabla_x\psi - \nabla_x\psi'| + |\Delta_x\psi||\psi - \psi'| + \|\rho'\|_{L^\infty}^{1/2}|\Delta_x\psi - \Delta_x\psi'|.$$

Furthermore, using (5.13) with $a = q - 2 > 1$ and (5.23) for ρ', we obtain

$$(iv) \lesssim_q (\|\rho\|_{L^\infty}^{q-5/2} + \|\rho'\|_{L^\infty}^{q-5/2})(\|\rho'\|_{L^\infty}^{1/2}|\Delta_x\psi'| + |\nabla_x\psi'|^2)|\psi - \psi'|.$$

Altogether, we get

$$|(III)| \lesssim_q \big(C_1|\nabla_x\psi|^2 + C_2|\nabla_x\psi||\nabla_x\psi'| + C_3|\nabla_x\psi'|^2 + C_4|\Delta_x\psi| + C_5|\Delta_x\psi'|\big)|\psi - \psi'|$$

$$+ (C_6|\nabla_x\psi| + C_7|\nabla_x\psi'|)|\nabla_x\psi - \nabla_x\psi'| + C_8|\Delta_x\psi - \Delta_x\psi'|, \tag{5.24}$$

where

$$C_1 = \|\rho\|_{L^\infty}^{q-5/2}(\|\rho\|_{L^\infty}^{1/2} + \|\rho'\|_{L^\infty}^{1/2}), \quad C_2 = \|\rho\|_{L^\infty}^{q-3}\|\rho'\|_{L^\infty},$$

$$C_3 = \|\rho'\|_{L^\infty}\big[\|\rho\|_{L^\infty}^{q-7/2}(1 + \|\rho\|_{L^\infty}) + \|\rho'\|_{L^\infty}^{q-7/2}(1 + \|\rho'\|_{L^\infty})\big],$$

$$C_4 = \|\rho\|_{L^\infty}^{q-3/2}(\|\rho\|_{L^\infty}^{1/2}\|\rho'\|_{L^\infty}^{1/2} + 1), \quad C_5 = \|\rho'\|_{L^\infty}(\|\rho\|_{L^\infty}^{q-5/2} + \|\rho'\|_{L^\infty}^{q-5/2}),$$

$$C_6 = \|\rho\|_{L^\infty}^{q-5/2}\|\rho'\|_{L^\infty}^{1/2}(\|\rho\|_{L^\infty}^{1/2} + \|\rho'\|_{L^\infty}^{1/2}), \quad C_7 = \|\rho\|_{L^\infty}^{q-3}\|\rho'\|_{L^\infty}^{1/2}(\|\rho\|_{L^\infty} + \|\rho'\|_{L^\infty}),$$

$$C_8 = \|\rho\|_{L^\infty}^{q-2}\|\rho'\|_{L^\infty}.$$

Conclusion of the proof of (5.18).

The function $\mathcal{L}$ can be split into terms and expressed as $\mathcal{L} = \mathcal{L}_0 + \mathcal{L}_{\mathrm{I}} + \mathcal{L}_{\mathrm{II}} + \mathcal{L}_{\mathrm{III}}$. As discussed at the start of this proof, $\mathcal{L}_0$ is the contribution of estimate (5.17) for $p = 2$. The other terms stem from (I), (II) and (III) in (5.20), and are obtained by one taking the L^2 norm in (5.21), (5.22) or (5.24), respectively. For instance, in the expression for $\mathcal{L}_{\mathrm{III}}$, all scalars C_i can be bounded with the use of (5.19). Likewise, the term $|\psi - \psi'|$ and the remaining factors involving C_1 and C_3 can be bounded with their H^2 norms. The other summands, and $\mathcal{L}_{\mathrm{I}}$ and $\mathcal{L}_{\mathrm{II}}$, can be estimated similarly, and this concludes the proof. $\quad\square$

Lemma 5.5 *(Lipschitz estimates for the nonlinearity). For $q \geq 7/2$ and any $\lambda \in \mathbb{R}$, there exists a polynomial function which vanishes at the origin $\mathscr{L}_{q,\lambda} : \mathbb{R}_0^+ \longrightarrow \mathbb{R}_0^+$ such that for all $\psi, \psi' \in \mathcal{B}_{\mathrm{el}}(\tau)$,*

$$\|V_{\mathrm{Hx}}[\rho]\psi - V_{\mathrm{Hx}}[\rho']\psi'\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))} \leq \mathscr{L}_{q,\lambda}(\alpha(\tau) + \|\psi^0\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})})\|\psi - \psi'\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))},$$
(5.25)

$$\|V_{\mathrm{Hx}}[\rho]\psi\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))} \leq (\alpha(\tau) + \|\psi^0\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})})\mathscr{L}_{q,\lambda}(\alpha(\tau) + \|\psi^0\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}).$$
(5.26)

Proof. By (5.3) in Lemma 5.1 and (5.18) in Lemma 5.4, we have for all $\psi, \psi' \in C^0([0,\tau]; H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))$,

$$\|V_{\mathrm{Hx}}[\rho]\psi - V_{\mathrm{Hx}}[\rho']\psi'\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))} \leq$$
$$\leq \mathscr{L}_{q,\lambda}(\max\{\|\psi\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))}, \|\psi'\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))}\})\|\psi - \psi'\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))}, \quad (5.27)$$

where $\mathscr{L}_{q,\lambda}$ is a polynomial by construction. Note that (5.25) follows from (5.27) by the definition of $\mathcal{B}_{\mathrm{el}}(\tau)$. In particular, (5.26) follows from (5.27) by our setting $\psi' \equiv 0$. $\quad\square$

6. Existence and uniqueness of nuclear configurations

In this section, we prove a local-in-time existence and uniqueness result for the Cauchy problem associated with (1.6) for given $\psi \in \mathcal{B}_{\mathrm{el}}(\tau)$ and $X(0) = X^0$ and $\dot{X}(0) = V(0)$, with $X^0, V^0 \in \mathbb{R}^{3N_{\mathrm{nuc}}}$ such that $X_K^0 \neq X_L^0$ for $1 \leq K \neq L \leq N_{\mathrm{nuc}}$.

Lemma 6.1. *Let $V^0 \in \mathbb{R}^{3N_{\mathrm{nuc}}}$ and $X^0 \in \mathbb{R}^{3N_{\mathrm{nuc}}}$ be given, with $X_K^0 \neq X_L^0$ for $K \neq L$.*

Then there exists $\tau > 0$ such that the following properties hold. For given $\psi \in \mathcal{B}_{\mathrm{el}}(\tau)$, the Cauchy problem associated with the system (1.6) with $X(0) = X^0$ and $\dot{X}(0) = V^0$ has a unique short-time solution $X \in \mathcal{B}_{\mathrm{nuc}}(\tau) \cap C^2([0,\tau]; B_\delta(X^0))$. The mapping

$$\mathcal{N} : \psi \in \mathcal{B}_{\mathrm{el}}(\tau) \longmapsto X \in \mathcal{B}_{\mathrm{nuc}}(\tau) \cap C^2([0,\tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})$$

is bounded with respect to the $C^1([0,\tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})$ topology, and is continuous as a map from $C^0([0,\tau]; L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))$ to $C^0([0,\tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})$.

Proof. *Part 1: Existence and uniqueness of X in $C^2([0,\tau]; B_\delta(X^0))$.*

Since ψ and so ρ are given, we write the acceleration function from (1.6) without parameters for now: $A = A(t, X)$. Note that t is an explicit variable for the A_K^1 terms, but not for the A_K^2 terms.

We define the compact set

$$\varkappa(\tau) := [0,\tau] \times B_\delta(X^0).$$

Note that we drop the dependence of this set on τ. By the triangle inequality, for all $X \in B_\delta(X^0)$ and $K = 1, \ldots, N_{\mathrm{nuc}}$,

$$|X_K| \le |X| \le |X^0| + |X - X^0| \le |X^0| + \delta(\tau). \tag{6.1}$$

First, we prove A is continuous in (t, X) on $\varkappa$. To this end, we pick a sequence $\{(t_n, X_n)\}_{n \in \mathbb{N}} \subset \varkappa$ with $(t_n, X_n) \xrightarrow{n \to \infty} (t^*, X) \in \varkappa$. The functions A_K^1 give for all n, with the use of the Cauchy–Schwarz and Hardy inequalities,

$$|A_K^1(t_n, X_n) - A_K^1(t^*, X_n)| \lesssim \sum_{k=1}^{N_{\mathrm{el}}} (|X_{nK} - \cdot|^{-2}, |(\psi_k(t_n, \cdot))^2 - (\psi_k(t^*, \cdot))^2|)_{L^2}$$

$$\lesssim \sum_{k=1}^{N_{\mathrm{el}}} \left(|X_{nK} - \cdot|^{-1} \max_{t \in [0,\tau]} |\psi_k(t, \cdot)|, |X_{nK} - \cdot|^{-1} |\psi_k(t_n, \cdot) - \psi_k(t^*, \cdot)| \right)_{L^2}$$

$$\lesssim \sum_{k=1}^{N_{\mathrm{el}}} \|\nabla_x \psi_k\|_{L^\infty([0,\tau];L^2(\mathbb{R}^3;\mathbb{C}^3))} \|\nabla_x \psi_k(t_n, \cdot) - \nabla_x \psi_k(t^*, \cdot)\|_{L^2(\mathbb{R}^3;\mathbb{C}^3)} \xrightarrow{n \to \infty} 0$$

as $\psi \in C^0([0,\tau]; H^1(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))$. Using this and Lemma 2.2, by which $A_K^1(t^*, \cdot) \in C^0(\mathbb{R}^{3N_{\mathrm{nuc}}}; \mathbb{C}^3)$, we get

$$|A_K^1(t_n, X_n) - A_K^1(t^*, X)| \le |A_K^1(t_n, X_n) - A_K^1(t^*, X_n)| + |A_K^1(t^*, X_n) - A_K^1(t^*, X)| \xrightarrow{n \to \infty} 0$$

for all n. The functions A_K^2 are not explicitly time-dependent, and are continuous on $B_\delta(X^0)$, and hence on $\varkappa$.

Since A is continuous on the compact set $\varkappa$, it is also uniformly bounded on $\varkappa$. By Lemma 2.2,

$$\|A_K^1\|_{C^0([0,\tau];W^{1,\infty}(B_\delta(X^0);\mathbb{C}^3))} \lesssim \|\psi\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))},$$

since $\psi \in \mathcal{B}_{\mathrm{el}}(\tau)$. The functions A_K^2 are bounded on $B_\delta(X^0)$ by

$$\|A_K^2\|_{L^\infty(B_\delta(X^0);\mathbb{C}^3)} \lesssim \sum_{L=1, L \ne K}^{N_{\mathrm{nuc}}} \left\| \frac{1}{|X_K - X_L|^2} \right\|_{L^\infty(B_\delta(X^0);\mathbb{C})}.$$

Furthermore, by Lemma 2.2, $A_K^1(t, \cdot)$ is uniformly Lipschitz continuous for all $t \in [0, \tau]$ and K, as

$$\|DA_K^1(t, \cdot)\|_{L^\infty(\mathbb{R}^3;\mathbb{C}^{3\times3})} \lesssim \sum_{k=1}^{N_{\mathrm{el}}} \|Df_K^{kk}(t, \cdot)\|_{L^\infty(\mathbb{R}^3;\mathbb{C}^{3\times3})} \lesssim \|\psi\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))},$$

since $\psi \in \mathcal{B}_{\mathrm{el}}(\tau)$. For the A_K^2 terms, we note that the functions $X \longmapsto (X_K - X_L)|X_K - X_L|^{-3}$ are locally Lipschitz on $B_\delta(X^0)$. Therefore, A is Lipschitz continuous in $X \in B_\delta(X^0)$ and uniformly in $t \in [0, \tau]$. We denote the corresponding Lipschitz constant by C_{L}, dropping its dependence on τ.

We now define $\mathcal{T}$ as the following mapping on the complete metric space $C^0([0,\tau]; B_\delta(X^0))$, equipped with the $C^0([0,\tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})$ norm:

$$\mathcal{T}[X](t) := X^0 + V^0 t + \int_0^t (t - \sigma) A(\sigma, X(\sigma)) \mathrm{d}\sigma. \tag{6.2}$$

By the boundedness of A,

$$\|\mathcal{T}[X] - X^0\|_{C^0([0,\tau];\mathbb{R}^{3N_{\mathrm{nuc}}})} \le |V^0|\tau + \frac{\tau^2}{2} \|A\|_{C^0(\varkappa;\mathbb{C}^{3N_{\mathrm{nuc}}})}$$

for all $X \in C^0([0, \tau]; B_\delta(X^0))$. Note that $\mathcal{T}$ maps $C^0([0, \tau]; B_\delta(X^0))$ into itself, as for $\tau > 0$ small enough it holds that

$$|V^0|\tau + \frac{\tau^2}{2}\|A\|_{C^0(\varkappa; \mathbb{C}^{3N_{\mathrm{nuc}}})} \leq \delta(\tau).$$

Hence, for all $X, X' \in C^0([0, \tau]; B_\delta(X^0))$,

$$\|\mathcal{T}[X] - \mathcal{T}[X']\|_{C^0([0, \tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})} \leq \max_{t \in [0, \tau]} \int_0^t (t - \sigma)|A(\sigma, X(\sigma)) - A(\sigma, X'(\sigma))|\mathrm{d}\sigma$$

$$\leq \frac{C_{\mathrm{L}}\tau^2}{2}\|X - X'\|_{C^0([0, \tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})}.$$

Note also that $\mathcal{T}$ is a strict contraction on $C^0([0, \tau]; B_\delta(X^0))$ in the $C^0([0, \tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})$ norm, as we can always shrink $\tau > 0$ so that

$$\frac{C_{\mathrm{L}}\tau^2}{2} < 1$$

holds. By the contraction mapping theorem, $\mathcal{T}$ has a unique fixed point in $C^0([0, \tau]; B_\delta(X^0))$. Because of this, (1.6) has a unique short-time solution in $C^2([0, \tau]; B_\delta(X^0))$.

Part 2: Localisation of X in $\mathcal{B}_{\mathrm{nuc}}(\tau)$.
   Integrating the ordinary differential equation in (1.6), we get

$$\|\dot{X}\|_{C^0([0, \tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})} \leq |V^0| + \tau\|A\|_{C^0(\varkappa; \mathbb{C}^{3N_{\mathrm{nuc}}})}.$$

Note that we can pick $\tau > 0$ small enough so that

$$\tau\|A\|_{C^0(\varkappa; \mathbb{C}^{3N_{\mathrm{nuc}}})} \leq 1$$

holds. Therefore, $X \in \mathcal{B}_{\mathrm{nuc}}(\tau) \cap C^2([0, \tau]; B_\delta(X^0))$.

Part 3: Boundedness and continuity of $\mathcal{N}$.
   From (6.1) with (3.1), $\mathcal{N}$ is bounded in the $C^1([0, \tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})$ norm:

$$\|X\|_{C^1([0, \tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})} \leq |X^0| + \delta(\tau) + \gamma.$$

To prove continuity of $\mathcal{N}$ in the $C^0([0, \tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})$ norm, we consider a sequence $\{\psi_n\}_{n \in \mathbb{N}} \subset \mathcal{B}_{\mathrm{el}}(\tau)$ such that $\psi_n \xrightarrow{n \to \infty} \psi \in \mathcal{B}_{\mathrm{el}}(\tau)$ in the $C^0([0, \tau]; L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))$ norm. Similarly to $X = \mathcal{N}[\psi]$, we define $X_n := \mathcal{N}[\psi_n]$ and $\rho_n := |\psi_n|^2$. Note that X and X_n are fixed points of the mapping $\mathcal{T}$ introduced in part 1 of the proof. Using this, we have for all $t \in [0, \tau]$,

$$|(X_n - X)(t)| \leq \int_0^t (t - \sigma)|A[\rho_n](X(\sigma)) - A[\rho](X(\sigma))|\mathrm{d}\sigma, \tag{6.3}$$

where

$$|A[\rho_n](X_n(\sigma)) - A[\rho](X(\sigma))| \le (\mathrm{I}) + (\mathrm{II}),$$

$$(\mathrm{I}) := \sum_{K=1}^{N_{\mathrm{nuc}}} |A_K^1[\rho_n](X_n(\sigma)) - A_K^1[\rho](X(\sigma))|,$$

$$(\mathrm{II}) := \sum_{K=1}^{N_{\mathrm{nuc}}} |A_K^2(X_n(\sigma)) - A_K^2(X(\sigma))|.$$

We further bound

$$(\mathrm{I}) \lesssim (\mathrm{Ia}) + (\mathrm{Ib}),$$

$$(\mathrm{Ia}) := \sum_{k=1}^{N_{\mathrm{el}}} \left| (\psi_k(t,\cdot), \Xi(\cdot - X_K)(\psi_{nk}(t,\cdot) - \psi_k(t,\cdot)))_{L^2} + (\psi_{nk}(t,\cdot) - \psi_k(t,\cdot), \Xi(\cdot - X_{nK})\psi_{nk}(t,\cdot))_{L^2} \right|,$$

$$(\mathrm{Ib}) := \sum_{k=1}^{N_{\mathrm{el}}} |(\psi_k(t,\cdot), \Xi(\cdot - X_{nK})\psi_{nk}(t,\cdot))_{L^2} - (\psi_k(t,\cdot), \Xi(\cdot - X_K)\psi_{nk}(t,\cdot))_{L^2}|,$$

where we use for short-hand notation the function $\Xi : \mathbb{R}^3 \longrightarrow \mathbb{R}^3$ (almost everywhere), $x \longmapsto x|x|^{-3}$. Arguing as in [8, p. 980], we state that (Ia) is bounded by

$$\beta_n := \sum_{k=1}^{N_{\mathrm{el}}} \sup_{(t,x) \in [0,\tau] \times \mathbb{R}^3} (|\cdot - x|^{-1}|\psi_k(t,\cdot) + \psi_{nk}(t,\cdot)|, |\cdot - x|^{-1}|\psi_{nk}(t,\cdot) - \psi_k(t,\cdot)|)_{L^2} \xrightarrow{n \longrightarrow \infty} 0,$$

as $\psi_n \xrightarrow{n \longrightarrow \infty} \psi$ in $C^0([0,\tau]; L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))$. We also have

$$(\mathrm{Ib}) \lesssim \sum_{k=1}^{N_{\mathrm{el}}} \|\nabla_x G[\psi_k, \psi_{nk}](X_{nK}) - \nabla_x G[\psi_k, \psi_{nk}](X_K)\|_{C^0([0,\tau]; \mathbb{C}^3)} \le C_{1,n}^{\mathrm{L}} |X_n - X|,$$

where G is as in (2.1), and where we used the fact that the functions $\nabla_x G[\psi_k, \psi_{nk}]$ are uniformly Lipschitz continuous in X for uniformly all $t \in [0,\tau]$. We note that (II) is also uniformly Lipschitz continuous, with some Lipschitz constant $C_{2,n}^{\mathrm{L}}$. For all n, $C_{1,n}^{\mathrm{L}}$ and $C_{2,n}^{\mathrm{L}}$ are uniformly bounded by C_{L}, since all ψ_n and ψ are taken from the bounded set $\mathcal{B}_{\mathrm{el}}(\tau)$. Altogether, from (6.3) we obtain

$$\|X_n - X\|_{C^0([0,\tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})} \lesssim \tau^2 \|X_n - X\|_{C^0([0,\tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})} + \tau^2 \beta_n.$$

It is then clear that for τ small enough the conclusion follows. $\quad\square$

7. Existence and uniqueness of electronic configurations

In this section, we prove a local-in-time existence and uniqueness result for the Cauchy problem associated with (1.4) for given $X \in \mathcal{B}_{\mathrm{nuc}}(\tau)$ and $\psi(0) = \psi^0 \in H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$.

Lemma 7.1. *Let $q \ge 7/2$ and $\lambda \in \mathbb{R}$. Let $\psi^0 \in H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$ be given. Then there exists $\tau > 0$ such that the following holds. For given $X \in \mathcal{B}_{\mathrm{nuc}}(\tau)$, the Cauchy problem associated with the system (1.4) with $\psi(0) = \psi^0$ has a unique short-time solution ψ in $\mathcal{B}_{\mathrm{el}}(\tau)$.*

Proof. The proof is based on Lemma 2.3, which ensures the existence and the $\mathcal{L}(H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))$ bounds of the propagator $U(t,s)$ for the family of linear Hamiltonians $\{H^{\mathrm{lin}}(t), t \in [0,\tau]\}$ from (2.7), and on Lemma 5.5, which ensures that the nonlinear mapping $\psi \longmapsto V_{\mathrm{Hx}}[\rho]\psi$ is locally Lipschitz in $H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$.

We define $\mathcal{F}$ as the following mapping on the complete metric space $C^0([0,\tau]; B_\alpha(\psi^0))$, equipped with the $C^0([0,\tau]; H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))$ norm:

$$\mathcal{F}[\psi](t) := U(t,0)\psi^0 - i \int_0^t U(t,\sigma)V_{\mathrm{Hx}}[\rho]\psi(\sigma)\mathrm{d}\sigma.$$

Note that we obtain for all $\psi \in C^0([0,\tau]; B_\alpha(\psi^0))$, using Lemma 2.3 (ii),

$$\mathcal{F}[\psi](0) = U(0,0)\psi^0 = \psi^0. \tag{7.1}$$

Note also that, provided

$$\left[1 + B_{\tau,\gamma} + \tau B_{\tau,\gamma}(2B_{\tau,\gamma} + 1)\mathscr{L}_{q,\lambda}(\alpha + \|\psi^0\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})})\right] \leq 2B_{\tau,\gamma}, \tag{7.2}$$

$\mathcal{F}$ maps the complete metric space $C^0([0,\tau]; B_\alpha(\psi^0))$ into itself as

$$\|\mathcal{F}[\psi] - \psi^0\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))} =$$

$$= \left\|[U(\cdot,0) - \mathrm{Id}]\psi^0 - i \int_0^{\cdot} U(\cdot,\sigma)V_{\mathrm{Hx}}[\rho]\psi(\sigma)\mathrm{d}\sigma\right\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))}$$

$$\leq B_{\tau,\gamma}\left(\|\psi^0\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} + \tau\|V_{\mathrm{Hx}}[\rho]\psi\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))}\right) + \|\psi^0\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \tag{7.3}$$

$$\overset{(5.26)}{\leq} \left[1 + B_{\tau,\gamma} + \tau B_{\tau,\gamma}(2B_{\tau,\gamma} + 1)\mathscr{L}_{q,\lambda}(\alpha + \|\psi^0\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})})\right]\|\psi^0\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}$$

$$\overset{(7.2)}{\leq} 2B_{\tau,\gamma}\|\psi^0\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} = \alpha,$$

where we used Lemma 2.3 (viii) in (7.3). Moreover, note that, provided

$$\tau B_{\tau,\gamma}\mathscr{L}_{q,\lambda}(\alpha(\tau) + \|\psi^0\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}) < 1, \tag{7.4}$$

$\mathcal{F}$ is a contraction on $C^0([0,\tau]; B_\alpha(\psi^0))$ in the $C^0([0,\tau]; H^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))$ norm, as for all $\psi, \psi' \in C^0([0,\tau]; B_\alpha(\psi^0))$,

$$\|\mathcal{F}[\psi] - \mathcal{F}[\psi']\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))} = \left\|\int_0^{\cdot} U(\cdot,\sigma)(V_{\mathrm{Hx}}[\rho]\psi(\sigma) - V_{\mathrm{Hx}}[\rho']\psi'(\sigma))\mathrm{d}\sigma\right\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))}$$

$$\leq \tau B_{\tau,\gamma}\|V_{\mathrm{Hx}}[\rho]\psi - V_{\mathrm{Hx}}[\rho']\psi'\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))} \tag{7.5}$$

$$\overset{(5.25)}{\leq} \tau B_{\tau,\gamma}\mathscr{L}_{q,\lambda}(\alpha + \|\psi^0\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})})\|\psi - \psi'\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))},$$

where we used Lemma 2.3 (viii) in (7.5). By the contraction mapping theorem, $\mathcal{F}$ has a unique fixed point in $C^0([0,\tau]; B_\alpha(\psi^0))$.

Note that we can always select $\tau > 0$ small enough such that the inequalities (7.2) and (7.4) are satisfied. Recall that $B_{\tau,\gamma}$ and α are of the form

$$B_{\tau,\gamma} = A_\gamma^{1+C_\gamma\tau}, \qquad \alpha(\tau) = 2A_\gamma^{1+C_\gamma\tau}\|\psi^0\|_{H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})},$$

with $A_\gamma, C_\gamma > 0$ as defined as in Lemma 2.3 (viii). If we pick A_γ and C_γ large, (7.2) is true for $\tau = 0$ and, by continuity, for $\tau > 0$ small enough.

It is now left to prove that this fixed point, denoted simply by ψ, is also of class $C^1([0,\tau]; L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))$; namely, it solves (1.4) strongly on $[0,\tau]$. To this end, we consider the following identity, which holds for all $0 \leq t < t' \leq \tau$:

$$i\frac{\psi(t') - \psi(t)}{t' - t} = (\mathrm{R}) := i\frac{U(t',0) - U(t,0)}{t' - t}\psi^0 + \int_0^t \frac{U(t',\sigma) - U(t,\sigma)}{t' - t}V_{\mathrm{Hx}}[\rho]\psi(\sigma)\mathrm{d}\sigma$$

$$+ \int_t^{t'} \frac{U(t',\sigma)}{t' - t}V_{\mathrm{Hx}}[\rho]\psi(\sigma)\mathrm{d}\sigma.$$

Further, we show that

$$\|(\mathrm{R}) - H[X(t),\rho]\psi(t)\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \xrightarrow{t' \longrightarrow t} 0.$$

This implies that $\psi(\cdot)$ is differentiable as a mapping $[0,\tau] \longmapsto L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$ such that

$$i\dot{\psi}(t) = H[X(t),\rho]\psi(t).$$

Note, in particular, that for a given $X \in \mathcal{B}_{\mathrm{nuc}}(\tau)$, $H[X(\cdot),\rho]\psi(\cdot)$ is a continuous mapping $[0,\tau] \longmapsto L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}})$, which implies that $\psi \in C^1([0,\tau]; L^2(\mathbb{R}^3; \mathbb{C}^{N_{\mathrm{el}}}))$. We bound

$$\|(\mathrm{R}) - H[X(t),\rho]\psi(t)\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \leq (\mathrm{I}) + (\mathrm{II}),$$

$$(\mathrm{I}) := \left\| i\frac{U(t',0) - U(t,0)}{t' - t}\psi^0 + \int_0^t \frac{U(t',\sigma) - U(t,\sigma)}{t' - t}V_{\mathrm{Hx}}[\rho]\psi(\sigma)\mathrm{d}\sigma - H^{\mathrm{lin}}(t)\psi(t) \right\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})},$$

$$(\mathrm{II}) := \left\| \int_t^{t'} \frac{U(t',\sigma)}{t' - t}V_{\mathrm{Hx}}[\rho]\psi(\sigma)\mathrm{d}\sigma - V_{\mathrm{Hx}}[\rho]\psi(t) \right\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}.$$

We get

$$\lim_{t' \longrightarrow t} (\mathrm{I}) = \left\| i\frac{\partial}{\partial t}[U(t,0)\psi^0] + \int_0^t \frac{\partial}{\partial t}[U(t,\sigma)V_{\mathrm{Hx}}[\rho]\psi(\sigma)]\mathrm{d}\sigma - H^{\mathrm{lin}}(t)\psi(t) \right\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}$$

$$= \left\| H^{\mathrm{lin}}(t)[U(t,0)\psi^0] + \int_0^t -iH^{\mathrm{lin}}(t)[U(t,\sigma)V_{\mathrm{Hx}}[\rho]\psi(\sigma)]\mathrm{d}\sigma - H^{\mathrm{lin}}(t)\psi(t) \right\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \quad (7.6)$$

$$= \|H^{\mathrm{lin}}(t)[\mathcal{F}[\psi(t)] - \psi(t)]\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} = 0, \quad (7.7)$$

where we used Lemma 2.3 (vii) (see also [31, Theorem 1.3 (6)]) in (7.6), the linearity of the Hamiltonians $H^{\mathrm{lin}}(t)$, and ψ being a fixed point of $\mathcal{F}$ in (7.7). On the other hand,

$$(\mathrm{II}) \leq (\mathrm{a}) + (\mathrm{b}),$$

$$(\mathrm{a}) := \left\| \frac{1}{t' - t}\int_t^{t'} U(t,\sigma)V_{\mathrm{Hx}}[\rho]\psi(\sigma)\mathrm{d}\sigma - V_{\mathrm{Hx}}[\rho]\psi(t) \right\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})},$$

$$(b) := \frac{1}{t' - t} \left\| \int_t^{t'} [U(t', \sigma) - U(t, \sigma)] V_{\text{Hx}}[\rho]\psi(\sigma) \mathrm{d}\sigma \right\|_{L^2(\mathbb{R}^3; \mathbb{C}^{N_{\text{el}}})}.$$

In the limit, (a) goes to zero, because of the fundamental theorem of calculus for Bochner integrals and Lemma 2.3 (ii). Moreover,

$$\lim_{t' \longrightarrow t} (b) \leq \lim_{t' \longrightarrow t} \frac{1}{t' - t} \int_t^{t'} \|[U(t', \sigma) - U(t, \sigma)] V_{\text{Hx}}[\rho]\psi(\sigma)\|_{L^2(\mathbb{R}^3; \mathbb{C}^{N_{\text{el}}})} \mathrm{d}\sigma$$

$$\leq \lim_{t' \longrightarrow t} \|[U(t', \cdot) - U(t, \cdot)] V_{\text{Hx}}[\rho]\psi\|_{C^0([0,T], L^2(\mathbb{R}^3; \mathbb{C}^{N_{\text{el}}}))} = 0, \tag{7.8}$$

where we used the uniform continuity of $U(t, s) V_{\text{Hx}}[\rho]\psi(s)$ on $[0, T]^2$ together with Lemma 2.3 (iv) in (7.8). Since ψ also is a fixed point of $\mathcal{F}$, by which $\psi(0) = \mathcal{F}[\psi](0) = \psi^0$ (see (7.1)), we know ψ is a strong solution to (1.4) on $[0, \tau]$.

We now show the uniqueness of the short-time solution ψ to (1.4) in the class $C^1([0, \tau]; L^2(\mathbb{R}^3; \mathbb{C}^{N_{\text{el}}})) \cap C^0([0, \tau]; B_\alpha(\psi^0))$: although the classical contraction mapping theorem also provides uniqueness, this is only in the class $C^0([0, \tau]; B_\alpha(\psi^0))$. So now we prove uniqueness in the different space $C^1([0, \tau]; L^2(\mathbb{R}^3; \mathbb{C}^{N_{\text{el}}}))$. To this end, we let ψ and ψ' be two short-time solutions to (1.4) in $C^1([0, \tau]; L^2(\mathbb{R}^3; \mathbb{C}^{N_{\text{el}}}))$. First, $(\psi - \psi')(0) = \psi^0 - \psi^0 = 0$. Moreover, for all $k = 1, \ldots, N_{\text{el}}$, using the partial differential equation in (1.4), we get

$$\frac{\mathrm{d}}{\mathrm{d}t}(\|\psi_k - \psi'_k\|_{L^2}^2) = \frac{\mathrm{d}}{\mathrm{d}t}((\psi_k - \psi'_k, \psi_k - \psi'_k)_{L^2})$$

$$= (\dot{\psi}_k - \dot{\psi}'_k, \psi_k - \psi'_k)_{L^2} + \overline{(\dot{\psi}_k - \dot{\psi}'_k, \psi_k - \psi'_k)_{L^2}} = (\text{I}) + (\text{II}),$$

where, using the fact that the linear Hamiltonians $H^{\text{lin}}(t)$ are self-adjoint on $L^2(\mathbb{R}^3; \mathbb{C}^{N_{\text{el}}})$, we have

$$(\text{I}) = i\left[(\psi_k - \psi'_k, (H^{\text{lin}}(t)(\psi - \psi'))_k)_{L^2} - ((H^{\text{lin}}(t)(\psi - \psi'))_k, \psi_k - \psi'_k)_{L^2}\right] = 0$$

and

$$(\text{II}) = i\left[\overline{((V_{\text{Hx}}[\rho]\psi - V_{\text{Hx}}[\rho']\psi')_k, \psi_k - \psi'_k)_{L^2}} - ((V_{\text{Hx}}[\rho]\psi - V_{\text{Hx}}[\rho']\psi')_k, \psi_k - \psi'_k)_{L^2}\right]$$

$$= 2\text{Im}((V_{\text{Hx}}[\rho]\psi - V_{\text{Hx}}[\rho']\psi')_k, \psi_k - \psi'_k)_{L^2}.$$

Using this, we get

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(\|\psi - \psi'\|_{L^2(\mathbb{R}^3; \mathbb{C}^{N_{\text{el}}})}^2\right) = \sum_{k=1}^{N_{\text{el}}} \frac{\mathrm{d}}{\mathrm{d}t}(\|\psi_k - \psi'_k\|_{L^2}^2)$$

$$= 2\text{Im}(V_{\text{Hx}}[\rho]\psi - V_{\text{Hx}}[\rho']\psi', \psi - \psi')_{L^2(\mathbb{R}^3; \mathbb{C}^{N_{\text{el}}})}$$

$$\leq C\|\psi - \psi'\|_{L^2(\mathbb{R}^3; \mathbb{C}^{N_{\text{el}}})}^2,$$

where $C = C(\|\psi\|_{C^0([0,\tau]; H^2(\mathbb{R}^3; \mathbb{C}^{N_{\text{el}}}))}, \|\psi'\|_{C^0([0,\tau]; H^2(\mathbb{R}^3; \mathbb{C}^{N_{\text{el}}}))}, \tau, q, \lambda, N_{\text{el}}) > 0$ stems from the Cauchy–Schwarz inequality and our combining (5.1) from Lemma 5.1 and (5.17) from Lemma 5.4. Finally, by Grönwall's lemma, we get $\psi = \psi'$, and this concludes the proof. $\square$

Lemma 7.2. *Let $q \geq 7/2$ and $\lambda \in \mathbb{R}$. Let $\tau > 0$ be such that the following holds: for given $X \in \mathcal{B}_{\text{nuc}}(\tau)$, $\psi \in \mathcal{B}_{\text{el}}(\tau)$ is the unique short-time solution to (1.4). Then the mapping*

$$\mathcal{E} : X \in \mathcal{B}_{\mathrm{nuc}}(\tau) \longmapsto \psi \in \mathcal{B}_{\mathrm{el}}(\tau)$$

is bounded and continuous as a map from $C^0\big([0,\tau];\mathbb{R}^{3N_{\mathrm{nuc}}}\big)$ to $C^0\big([0,\tau];L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})\big)$.

Proof. Since $\mathcal{B}_{\mathrm{el}}(\tau)$ is a bounded subset of $C^0([0,\tau];L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))$, the mapping $\mathcal{E}$ is bounded in the $C^0([0,\tau];L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))$ norm. To prove continuity of $\mathcal{E}$ as a map from $C^0([0,\tau];\mathbb{R}^{3N_{\mathrm{nuc}}})$ to $C^0\big([0,\tau];L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})\big)$, we consider a sequence $\{X_n\}_{n\in\mathbb{N}} \subset \mathcal{B}_{\mathrm{nuc}}(\tau)$ such that $X_n \xrightarrow{n\longrightarrow\infty} X \in \mathcal{B}_{\mathrm{nuc}}(\tau)$ in the $C^0([0,\tau];\mathbb{R}^{3N_{\mathrm{nuc}}})$ norm. Similarly to $\psi = \mathcal{E}[X]$, we define $\psi_n := \mathcal{E}[X_n]$ with $\rho_n := |\psi_n|^2$. Then

$$i\frac{\partial}{\partial t}(\psi_n - \psi) = H[X,\rho](\psi_n - \psi) + \zeta_n, \qquad (\psi_n - \psi)(0) = 0,$$

with

$$\begin{aligned}
\zeta_n &:= \zeta_n^1 + \zeta_n^2 + \zeta_n^3,\\
\zeta_n^1 &:= V_{\mathrm{ext}}[X_n]\psi_n - V_{\mathrm{ext}}[X]\psi - V_{\mathrm{ext}}[X](\psi_n - \psi) = (V_{\mathrm{ext}}[X_n] - V_{\mathrm{ext}}[X])\psi_n,\\
\zeta_n^2 &:= V_{\mathrm{H}}[\rho_n]\psi_n - V_{\mathrm{H}}[\rho]\psi - V_{\mathrm{H}}[\rho](\psi_n - \psi)\\
&= \sum_{k=1}^{N_{\mathrm{el}}}\{\mathrm{Re}\big[\overline{(\psi_{nk} - \psi_k)}(\psi_{nk} + \psi_k)\big] * |\cdot|^{-1}\}\psi_n, \qquad\qquad (7.9)\\
\zeta_n^3 &:= V_{\mathrm{x}}[\rho_n]\psi_n - V_{\mathrm{x}}[\rho]\psi - V_{\mathrm{x}}[\rho](\psi_n - \psi) = \lambda[\rho_n^{q-1} - \rho^{q-1}]\psi_n,
\end{aligned}$$

where we used $|a|^2 - |b|^2 = \mathrm{Re}[\overline{(a-b)}(a+b)]$ in (7.9). We denote by $\{H[X(t),\rho], t \in [0,T]\}$ the family of KS Hamiltonians for the given $X \in \mathcal{B}_{\mathrm{nuc}}(\tau)$. Note that since ψ and thus ρ are fixed now, these Hamiltonians are acting linearly on $\psi_n - \psi$, and can thus be written, similarly to (2.7), as

$$H(t) = -\tfrac{1}{2}\Delta_x + V(t) + V_{\mathrm{Hx}}[\rho]$$

with V from (2.8). The linear potential $V(t) + V_{\mathrm{Hx}}[\rho]$ satisfies Assumption (A.1) in [31, Theorem 1.1]; hence, there exists a family of evolution operators $\{\mathfrak{U}(t,s),(t,s) \in [0,T]^2\}$, associated with this family of Hamiltonians, satisfying properties (i)–(iv) of Lemma 2.3. By this, from which it follows that for fixed $t \in [0,T]$, $\mathfrak{U}(t,\cdot)\zeta_n \in C^0([0,t],L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))$, and [31, Corollary 1.2 (4)], we can argue as in [8, p. 982], and the corresponding integral representation holds for all $t \in [0,T]$:

$$(\psi_n - \psi)(t) = -i\int_0^t \mathfrak{U}(t,\sigma)\zeta_n(\sigma)\mathrm{d}\sigma.$$

Using Lemma 2.3 (iii), we bound for all $n \in \mathbb{N}$ and $t \in [0,\tau]$

$$\|(\psi_n - \psi)(t)\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \lesssim \sum_{j\in\{1,2,3\}}\int_0^t \|\zeta_n^j(\sigma)\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}\mathrm{d}\sigma.$$

So now we deduce $L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})$ estimates on $\zeta_n^j(\sigma)$ for $j \in \{1,2,3\}$ for all $\sigma \in (0,t)$, using the fact that ψ_n and ψ are elements in $\mathcal{B}_{\mathrm{el}}(\tau)$, which makes them uniformly bounded with respect to n in $C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))$. For $j = 1$, as noted in [8, p. 982], it holds for all $0 < \sigma < t \le \tau \le T$ that

$$\|\zeta_n^1(\sigma)\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \le C_{1,n} \xrightarrow{n\longrightarrow\infty} 0$$

for some $C_{1,n} = C_{1,n}(\alpha, \psi^0) > 0$. For $j = 2$, we use the mapping G from (2.1). This gives for all $\sigma \in (0, t)$,

$$\|\zeta_n^2(\sigma)\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \leq \sum_{k=1}^{N_{\mathrm{el}}} \|G[\psi_{nk}(\sigma) - \psi_k(\sigma), \psi_{nk}(\sigma) + \psi_k(\sigma)]\|_{L^\infty} \|\psi_n(\sigma)\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}$$

$$\lesssim \sum_{k=1}^{N_{\mathrm{el}}} \|\psi_{nk}(\sigma) - \psi_k(\sigma)\|_{L^2} \|\psi_{nk}(\sigma) + \psi_k(\sigma)\|_{H^2} \|\psi_n\|_{L^\infty([0,\tau];L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))}$$

$$\leq C_2 \|\psi_n(\sigma) - \psi(\sigma)\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}$$

for some $C_2 = C_2(\alpha, \psi^0) > 0$. For $j = 3$ and all $\sigma \in (0, t)$,

$$\|\zeta_n^3(\sigma)\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \overset{(5.13)}{\lesssim}_{q,\lambda} (\|\rho_n(\sigma)\|_{L^\infty}^{q-3/2} + \|\rho(\sigma)\|_{L^\infty}^{q-3/2}) \|\rho_n(\sigma)\|_{L^\infty}^{1/2} \|\psi_n(\sigma) - \psi(\sigma)\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}$$

$$\overset{(5.19)}{\leq} C_3 \|\psi_n(\sigma) - \psi(\sigma)\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}$$

for some $C_3 = C_3(q, \alpha, \psi^0) > 0$. Combining these three estimates, we have for all $t \in [0, \tau]$,

$$\|(\psi_n - \psi)(t)\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \leq C_{1,n}\tau + (C_2 + C_3) \int_0^t \|(\psi_n - \psi)(\sigma)\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \mathrm{d}\sigma.$$

By Grönwall's lemma, we conclude that for all $t \in [0, \tau]$,

$$\|(\psi_n - \psi)(t)\|_{L^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \leq C_{1,n}\tau e^{(C_2+C_3)t},$$

and this concludes the proof. $\square$

8. Proof of Theorem 1.1

In this section, we prove the main result, Theorem 1.1.

Lemma 8.1. *Let $q \geq 7/2$ and $\lambda \in \mathbb{R}$. Further, let $\psi^0 \in H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})$, $V^0 \in \mathbb{R}^{3N_{\mathrm{nuc}}}$ and $X^0 \in \mathbb{R}^{3N_{\mathrm{nuc}}}$ be given, with $X_K^0 \neq X_L^0$ for $K \neq L$.*
Then there exists $\tau > 0$ such that the initial-value problem associated with the system (1.1) with $\psi(0) = \psi^0$, $X(0) = X^0$ and $\dot{X}(0) = V^0$ has a solution $(\psi, X) \in \mathcal{X}(\tau)$.

Proof. Let $\tau > 0$ be such that the following statements hold. For given $\psi \in \mathcal{B}_{\mathrm{el}}(\tau)$, (1.6) has a unique solution $X \in \mathcal{B}_{\mathrm{nuc}}(\tau) \cap C^2([0, \tau]; B_\delta(X^0))$, and for given $X \in \mathcal{B}_{\mathrm{nuc}}(\tau)$, (1.4) has a unique solution $\psi \in \mathcal{B}_{\mathrm{el}}(\tau)$. The existence of such τ was proven in Lemmas 6.1 and 7.1. We define the inclusion

$$\mathcal{I} : \mathcal{B}_{\mathrm{nuc}}(\tau) \cap C^2([0, \tau]; \mathbb{R}^{3N_{\mathrm{nuc}}}) \hookrightarrow \mathcal{B}_{\mathrm{nuc}}(\tau),$$

which is a continuous and compact mapping. Also, we define the mapping

$$\mathcal{K} : \mathcal{B}_{\mathrm{nuc}}(\tau) \longrightarrow \mathcal{B}_{\mathrm{nuc}}(\tau), \qquad \mathcal{K} := \mathcal{I} \circ \mathcal{N} \circ \mathcal{E}.$$

Since by Lemma 6.1, $\mathcal{N}$ is bounded in the $C^1([0, \tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})$ topology, by the Arzelà–Ascoli theorem it follows that $\mathcal{K}$ is a compact mapping, where $\mathcal{B}_{\mathrm{nuc}}(\tau)$ is equipped with the $C^0([0, \tau]; \mathbb{R}^{3N_{\mathrm{nuc}}})$ topology.

By the classical Schauder fixed-point theorem, $\mathcal{K}$ has a fixed point X in $\mathcal{B}_{\mathrm{nuc}}(\tau)$. If we set $\psi := \mathcal{E}[X]$, the corresponding pair (ψ, X) is the desired solution, and this concludes the proof. $\square$

Lemma 8.2. *Let $q \geq 7/2$ and $\lambda \in \mathbb{R}$. Let $(X, \psi), (X', \psi') \in \mathcal{X}(\tau)$ be two solutions to (1.1) for some $\tau > 0$. Then, for all $t \in [0, \tau]$,*

$$|(\ddot{X} - \ddot{X}')(t)| \leq C[|(X - X')(t)| + \|(\psi - \psi')(t)\|_{L^{3,\infty}(\mathbb{R}^3;\mathbb{C}^{N_{el}})}], \tag{8.1}$$

$$\|(\psi - \psi')(t)\|_{L^{3,\infty}(\mathbb{R}^3;\mathbb{C}^{N_{el}})} \leq C \int_0^t \frac{1}{\sqrt{t - \sigma}}[|(X - X')(\sigma)| + \|(\psi - \psi')(\sigma)\|_{L^{3,\infty}(\mathbb{R}^3;\mathbb{C}^{N_{el}})}]d\sigma, \tag{8.2}$$

where $C = C(\|\psi\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{el}}))}, \|\psi'\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{el}}))})$.

Proof. We focus on justifying each estimate separately, as follows.

Proof of (8.1). In this proof, we use for short-hand notation the function $\Xi : \mathbb{R}^3 \longrightarrow \mathbb{R}^3$ (almost everywhere), $x \longmapsto x|x|^{-3}$ again.

For all $t \in [0, \tau]$ and $K = 1, \ldots, N_{nuc}$,

$$|(\ddot{X}_K - \ddot{X}'_K)(t)| \leq |A^1_K[\rho(t)](X(t)) - A^1_K[\rho'(t)](X'(t))| + |A^2_K(X(t)) - A^2_K(X'(t))|$$

$$\leq (\mathrm{I}) + (\mathrm{II}) + (\mathrm{III}),$$

$$(\mathrm{I}) := |A^1_K[\rho(t)](X(t)) - A^1_K[\rho(t)](X'(t))|,$$

$$(\mathrm{II}) := |A^1_K[\rho(t)](X'(t)) - A^1_K[\rho'(t)](X'(t))|,$$

$$(\mathrm{III}) := |A^2_K(X(t)) - A^2_K(X'(t))|.$$

By Lemma 2.2 on the force functions, $A^1_K[\rho]$ are uniformly Lipschitz continuous in the nuclear variable for all $t \in [0, \tau]$ and K, by which

$$(\mathrm{I}) \lesssim \sum_{k=1}^{N_{el}} |(\psi_k(t), \Xi(\cdot - X_K(t))\psi_k(t))_{L^2} - (\psi_k(t), \Xi(\cdot - X'_K(t))\psi_k(t))_{L^2}|$$

$$\overset{(2.6)}{\leq} C_{\mathrm{I}}|(X_K - X'_K)(t)| \leq C_{\mathrm{I}}|(X - X')(t)|$$

for some $C_{\mathrm{I}} = C_{\mathrm{I}}(\|\psi\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{el}}))}) > 0$. Also,

$$(\mathrm{II}) \leq C_{\mathrm{II}}\|(\psi - \psi')(t)\|_{L^{3,\infty}(\mathbb{R}^3;\mathbb{C}^{N_{el}})}$$

for some $C_{\mathrm{II}} = C_{\mathrm{II}}(\|\psi\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{el}}))}, \|\psi'\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{el}}))}) > 0$ by Hölder's inequality in Lorentz spaces (A.1) and the fact that $\|\cdot\|^{-2}_{\mathbb{R}^3} \in L^{3/2,\infty}$. Since $X, X' \in \mathcal{B}_{nuc}(\tau)$, we bound (III) similarly to the bounding in part 1 of the proof of Lemma 6.1:

$$(\mathrm{III}) \lesssim \sum_{L=1, L \neq K}^{N_{nuc}} |\Xi((X_K - X_L)(t)) - \Xi((X'_K - X'_L)(t))| \lesssim_{\delta, X^0} |(X - X')(t)|.$$

Since these results hold for all K, (8.1) follows.

Proof of (8.2). Similarly to the proof of Lemma 7.1,

$$i\frac{\partial}{\partial t}(\psi - \psi') = -\tfrac{1}{2}\Delta_x(\psi - \psi') + V_{ext}[X](\psi - \psi') + V_{Hx}[\rho](\psi - \psi') + \widetilde{\zeta},$$

$$(\psi - \psi')(0) = 0,$$

where $\widetilde{\zeta} := \widetilde{\zeta}^1 + \widetilde{\zeta}^2 + \widetilde{\zeta}^3$, with for $j \in \{1, 2, 3\}$, $\widetilde{\zeta}^j$ being ζ_n^j with $(X_n, \psi_n) \longmapsto (X', \psi')$. As the operator $-\Delta_x/2$ generates the free propagator U_0, we write the equivalent integral equation for all $t \in [0, \tau]$:

$$(\psi - \psi')(t) = -i \int_0^t U_0(t - \sigma)\{V_{\text{ext}}[X(\sigma)](\psi - \psi')(\sigma) + V_{\text{Hx}}[\rho](\psi - \psi')(\sigma) + \widetilde{\zeta}(\sigma)\}d\sigma.$$

We recall that by Lemma 6 in [8], for all $\sigma \in (0, \tau]$ and $f \in L^{3/2, \infty}$, it holds that

$$\|U_0(\sigma)f\|_{L^{3,\infty}} \lesssim \frac{1}{\sqrt{\sigma}}\|f\|_{L^{3/2,\infty}}.$$

Using this estimate, we obtain for all $t \in [0, \tau]$ and $k = 1, \dots, N_{\text{el}}$,

$$\|(\psi_k - \psi_k')(t)\|_{L^{3,\infty}} \lesssim \int_0^t \frac{1}{\sqrt{t - \sigma}}\Big[\|(V_{\text{ext}}[X(\sigma)](\psi - \psi'))_k(\sigma)\|_{L^{3/2,\infty}}$$

$$+ \|(V_{\text{H}}[\rho](\psi - \psi'))_k(\sigma)\|_{L^{3/2,\infty}} + \|(V_{\text{x}}[\rho](\psi - \psi'))_k(\sigma)\|_{L^{3/2,\infty}}$$

$$+ \sum_{j \in \{1,2,3\}} \|(\widetilde{\zeta}^j(\sigma))_k\|_{L^{3/2,\infty}}\Big]d\sigma.$$

Since $\|\cdot\|_{\mathbb{R}^3}^{-1} \in L^{3,\infty}$, by Hölder's inequality on $L^{3/2,\infty}$, we obtain for all $\sigma \in (0, t)$ and $k = 1, \dots, N_{\text{el}}$,

$$\|(V_{\text{ext}}[X(\sigma)](\psi - \psi'))_k(\sigma)\|_{L^{3/2,\infty}} \lesssim \sum_{K=1}^{N_{\text{nuc}}} \||\cdot - X_K(\sigma)|^{-1}\|_{L^{3,\infty}}\|(\psi_k - \psi_k')(\sigma)\|_{L^{3,\infty}}$$

$$\lesssim \||\cdot|^{-1}\|_{L^{3,\infty}}\|(\psi_k - \psi_k')(\sigma)\|_{L^{3,\infty}}$$

$$\lesssim \|(\psi_k - \psi_k')(\sigma)\|_{L^{3,\infty}}.$$

Note also that

$$\|(V_{\text{H}}[\rho](\psi - \psi'))_k(\sigma)\|_{L^{3/2,\infty}} \lesssim \|\rho(\sigma) * |\cdot|^{-1}\|_{L^{3,\infty}}\|(\psi_k - \psi_k')(\sigma)\|_{L^{3,\infty}} \tag{8.3}$$

$$\lesssim \|\rho(\sigma)\|_{L^1}\||\cdot|^{-1}\|_{L^{3,\infty}}\|(\psi_k - \psi_k')(\sigma)\|_{L^{3,\infty}} \tag{8.4}$$

$$\leq C_{\text{H}}\|(\psi_k - \psi_k')(\sigma)\|_{L^{3,\infty}}$$

for some $C_{\text{H}} = C_{\text{H}}(\|\psi\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\text{el}}}))}) > 0$. Here we used Hölder's inequality on $L^{3/2,\infty}$ in (8.3) and Young's convolution inequality on $L^{3,\infty}$ in (8.4). Moreover,

$$\|(V_{\text{x}}[\rho](\psi - \psi'))_k(\sigma)\|_{L^{3/2,\infty}} \lesssim_\lambda \|[\rho(\sigma)]^{q-1}\|_{L^{3,\infty}}\|(\psi_k - \psi_k')(\sigma)\|_{L^{3,\infty}} \tag{8.5}$$

$$\lesssim \|[\rho(\sigma)]^{q-1}\|_{L^3}\|(\psi_k - \psi_k')(\sigma)\|_{L^{3,\infty}} \tag{8.6}$$

$$\leq C_{\text{x}}\|(\psi_k - \psi_k')(\sigma)\|_{L^{3,\infty}} \tag{8.7}$$

for some $C_{\text{x}} = C_{\text{x}}(\|\psi\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\text{el}}}))}, q) > 0$. Here we used Hölder's inequality on $L^{3/2,\infty}$ in (8.5) and Proposition 4.2 from [4] in (8.6). In (8.7), we used Sobolev's inequality with interpolation, and the embedding of H^2 into L^∞, by which, with $\theta := 6(q - 1) > 6$,

$$\|[\rho(\sigma)]^{q-1}\|_{L^3}^3 \lesssim_q \sum_{k=1}^{N_{\mathrm{el}}} \|[\psi_k(\sigma)]^{(\theta-6)+6}\|_{L^1}$$

$$\lesssim \sum_{k=1}^{N_{\mathrm{el}}} \|\psi_k(\sigma)\|_{L^\infty}^{\theta-6} \|\psi_k(\sigma)\|_{L^6}^6 \lesssim \sum_{k=1}^{N_{\mathrm{el}}} \|\psi_k\|_{C^0([0,\tau];H^2)}^\theta.$$

On the other hand,

$$\|(\widetilde{\zeta}^1(\sigma))_k\|_{L^{3/2,\infty}} \lesssim \sum_{K=1}^{N_{\mathrm{nuc}}} \|(|\cdot-X_K(\sigma)|^{-1} - |\cdot-X_K'(\sigma)|^{-1})\psi_k'(\sigma,\cdot)\|_{L^{3/2,\infty}}$$

$$= \sum_{K=1}^{N_{\mathrm{nuc}}} \|(|\cdot-(X_K-X_K')(\sigma)|^{-1} - |\cdot|^{-1})\psi_k'(\sigma,\cdot+X_K')\|_{L^{3/2,\infty}}$$

$$\lesssim \|\psi_k'(\sigma)\|_{L^\infty} \sum_{K=1}^{N_{\mathrm{nuc}}} \||\cdot|^{-1}|\cdot-(X_K-X_K')(\sigma)|^{-1}\|_{L^{3/2,\infty}}|(X_K-X_K')(\sigma)| \qquad (8.8)$$

$$\lesssim \|\psi_k'\|_{C^0([0,\tau];H^2)} \||\cdot|^{-1}\|_{L^{3,\infty}} \sum_{K=1}^{N_{\mathrm{nuc}}} \||\cdot-(X_K-X_K')(\sigma)|^{-1}\|_{L^{3,\infty}}|(X_K-X_K')(\sigma)| \quad (8.9)$$

$$\lesssim_{N_{\mathrm{nuc}}} \|\psi_k'\|_{C^0([0,\tau];H^2)} \||\cdot|^{-1}\|_{L^{3,\infty}}^2 |(X-X')(\sigma)|$$

$$\lesssim \|\psi_k'\|_{C^0([0,\tau];H^2)} |(X-X')(\sigma)|,$$

where we used the triangle inequality written as $||\cdot|-|\cdot-(X_K-X_K')(\sigma)|| \le |(X_K-X_K')(\sigma)|$ in (8.8), and Hölder's inequality in $L^{3/2,\infty}$ and the embedding of H^2 into L^∞ in (8.9). Moreover,

$$\|(\widetilde{\zeta}^2(\sigma))_k\|_{L^{3/2,\infty}} \lesssim \sum_{\ell=1}^{N_{\mathrm{el}}} \|\{[\overline{(\psi_\ell-\psi_\ell')(\sigma)}(\psi_\ell+\psi_\ell')(\sigma)]*|\cdot|^{-1}\}\psi_k'(\sigma)\|_{L^{3/2,\infty}}$$

$$\lesssim \sum_{\ell=1}^{N_{\mathrm{el}}} \|[\overline{(\psi_\ell-\psi_\ell')(\sigma)}(\psi_\ell+\psi_\ell')(\sigma)]*|\cdot|^{-1}\|_{L^{6,\infty}} \|\psi_k'(\sigma)\|_{L^{2,\infty}} \qquad (8.10)$$

$$\lesssim \sum_{\ell=1}^{N_{\mathrm{el}}} \|[\overline{(\psi_\ell-\psi_\ell')(\sigma)}(\psi_\ell+\psi_\ell')(\sigma)]*|\cdot|^{-1}\|_{L^{6,2}} \|\psi_k'(\sigma)\|_{L^2} \qquad (8.11)$$

$$\lesssim \|\psi_k'\|_{C^0([0,\tau];H^2)} \sum_{\ell=1}^{N_{\mathrm{el}}} \|\overline{(\psi_\ell-\psi_\ell')(\sigma)}(\psi_\ell+\psi_\ell')(\sigma)\|_{L^{6/5,2}} \||\cdot|^{-1}\|_{L^{3,\infty}} \qquad (8.12)$$

$$\lesssim \|\psi_k'\|_{C^0([0,\tau];H^2)} \sum_{\ell=1}^{N_{\mathrm{el}}} [\|\psi_\ell(\sigma)\|_{L^2} + \|\psi_\ell'(\sigma)\|_{L^2}] \|(\psi_\ell-\psi_\ell')(\sigma)\|_{L^{3,\infty}} \qquad (8.13)$$

$$\le C_2 \|(\psi-\psi')(\sigma)\|_{L^{3,\infty}(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}$$

for some $C_2 = C_2(\|\psi\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))}, \|\psi'\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))}) > 0$. Here we used Hölder's inequality on $L^{3/2,\infty}$ in (8.10), Proposition 4.2 from [4] in (8.11), Young's convolution inequality on $L^{6,2}$ in (8.12) and Hölder's inequality on $L^{6/5,2}$ in (8.13). Finally,

$$\|(\widetilde{\zeta}^3(\sigma))_k\|_{L^{3/2,\infty}} \overset{(5.13)}{\lesssim}_{q,\lambda} \|[\rho(\sigma)]^{q-3/2} + [\rho'(\sigma)]^{q-3/2}\|_{L^\infty} \|\psi_k'(\sigma)|\psi(\sigma)-\psi'(\sigma)|\|_{L^{3/2,\infty}}$$

$$\lesssim [\|\rho(\sigma)\|_{L^\infty}^{q-3/2} + \|\rho'(\sigma)\|_{L^\infty}^{q-3/2}] \|\psi_k'(\sigma)\|_{L^{3,\infty}} \||(\psi-\psi')(\sigma)|\|_{L^{3,\infty}(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \qquad (8.14)$$

$$\lesssim_q \Big[\sum_{\ell'=1}^{N_{\mathrm{el}}} \|\psi_{\ell'}(\sigma)\|_{H^2}^{2q-3} + \sum_{k'=1}^{N_{\mathrm{el}}} \|\psi'_{k'}(\sigma)\|_{H^2}^{2q-3} \Big] \|\psi'_k(\sigma)\|_{L^3} \sum_{\ell=1}^{N_{\mathrm{el}}} \|(\psi_\ell - \psi'_\ell)(\sigma)\|_{L^{3,\infty}} \quad (8.15)$$

$$\leq C_3 \|(\psi - \psi')(\sigma)\|_{L^{3,\infty}(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})} \tag{8.16}$$

for some $C_3 = C_3(\|\psi\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))}, \|\psi'\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))}, q) > 0$. Here we used Hölder's inequality on $L^{3/2,\infty}$ in (8.14), (5.19) and Proposition 4.2 from [4] in (8.15), and Sobolev's embedding theorem with interpolation in (8.16). Since all these estimates hold for all $\sigma \in (0,t)$, and $k = 1, \ldots, N_{\mathrm{el}}$, (8.2) follows. $\quad\square$

Proof of Theorem 1.1. Let $\tau > 0$ be such that the following statements hold. For given $\psi \in \mathcal{B}_{\mathrm{el}}(\tau)$, (1.6) has a unique solution $X \in \mathcal{B}_{\mathrm{nuc}}(\tau) \cap C^2([0,\tau]; B_\delta(X^0))$, and for given $X \in \mathcal{B}_{\mathrm{nuc}}(\tau)$, (1.4) has a unique solution $\psi \in \mathcal{B}_{\mathrm{el}}(\tau)$. The existence of such τ was proven in Lemmas 6.1 and 7.1. The existence of the solution $(X, \psi) \in \mathcal{X}(\tau)$ to (1.1) was proven in Lemma 8.1. Uniqueness of this solution follows from Lemma 8.2. For two solutions $(X, \psi), (X', \psi') \in \mathcal{X}(\tau)$ and $p > 2$, we define the function $h \in C^0([0,\tau]; \mathbb{R}_0^+)$ by

$$h(t) := [|(X - X')(t)| + \|(\psi - \psi')(t)\|_{L^{3,\infty}(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}]^p.$$

Since X and X' both solve (1.6) on $[0,\tau]$ and thus are fixed points of the mapping $\mathcal{T}$ in (6.2), for all $t \in [0,\tau]$,

$$|(X - X')(t)| \leq \int_0^t (t - \sigma)|(\ddot{X} - \ddot{X}')(\sigma)|\mathrm{d}\sigma.$$

Using this in combination with Lemma 8.2 in (8.17) and Hölder's inequality, we have for all $t \in [0,\tau]$,

$$h(t) \lesssim_p C\left\{ \int_0^t \Big(t - \sigma + \frac{1}{\sqrt{t-\sigma}}\Big)[|(X - X')(\sigma)| + \|(\psi - \psi')(\sigma)\|_{L^{3,\infty}(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}})}]\mathrm{d}\sigma \right\}^p \tag{8.17}$$

$$\lesssim C\Big\|\Big(t - \cdot + \frac{1}{\sqrt{t-\cdot}}\Big)h^{1/p}\Big\|_{L^1([0,t];\mathbb{R})}^p$$

$$\lesssim C\Big\|t - \cdot + \frac{1}{\sqrt{t-\cdot}}\Big\|_{L^{p'}([0,t];\mathbb{R})}^p \|h^{1/p}\|_{L^p([0,t];\mathbb{R})}^p \lesssim_\tau C\int_0^t h(\sigma)\mathrm{d}\sigma,$$

where $C = C(\|\psi\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))}, \|\psi'\|_{C^0([0,\tau];H^2(\mathbb{R}^3;\mathbb{C}^{N_{\mathrm{el}}}))})$ is from Lemma 8.2. Using Grönwall's inequality, we obtain $h \leq 0$ on $[0,\tau]$. Since $h \geq 0$ too by definition, and $h(0) = 0$ since $X(0) = X'(0) = X^0$ and $\psi(0) = \psi'(0) = \psi^0$, we get $h \equiv 0$, by which $(X, \psi) = (X', \psi')$. This completes the proof. $\quad\square$

CRediT authorship contribution statement

Björn Baumeier: Conceptualization, Funding acquisition, Supervision, Writing – review. **Onur Çaylak:** Conceptualization. **Carlo Mercuri:** Conceptualization, Formal analysis, Funding acquisition, Methodology, Supervision, Writing – original draft, Writing – review & editing. **Mark Peletier:** Conceptualization, Methodology, Writing – review. **Georg Prokert:** Conceptualization, Writing – review (in part). **Wouter Scharpach:** Conceptualization, Formal analysis, Project administration, Writing – original draft, Writing – review & editing.

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Appendix A. Notation

Throughout this article, we use the following notation:

- We use $A \lesssim B$ to denote $|A| \leq CB$, where $0 < C < \infty$.
- We use $A \lesssim_{\alpha,\beta} B$ to denote dependence on parameters α and β: so $|A| \leq C_{\alpha,\beta}B$ with $0 < C_{\alpha,\beta} < \infty$. In our notation, $A \lesssim_\alpha B \lesssim_\beta \Gamma$ means that $A \lesssim_{\alpha,\beta} \Gamma$.

Further, we use the following normed spaces:

- We use the notation $L^{p,r}(\mathbb{R}^3) = L^{p,r}$, $p \in [1,\infty)$, $r \in [1,\infty]$, for Lorentz spaces, with
  - the radial nonincreasing rearrangement of measurable functions f on $\mathbb{R}^3$

  $$f^*(t) = \inf\{s > 0 \,||\, \{x \in \mathbb{R}^3 \,||\, |f(x)| > s\}| \leq t\},$$

  - the average of f^*

  $$f^{**}(t) = \frac{1}{t}\int_0^t f^*(s)\mathrm{d}s,$$

  - the norms

  $$\|f\|_{L^{p,r}}^r = \int_0^\infty |t^{1/p}f^{**}(t)|^r \frac{\mathrm{d}t}{t}$$

  on $L^{p,r}$, $r < \infty$, and

  $$\|f\|_{L^{p,\infty}} = \sup_{t\in\mathbb{R}} |t^{1/p}f^{**}(t)|$$

  on the weak Lebesgue spaces $L^{p,\infty}$.
- We use the notation $W^{k,p}(\mathbb{R}^3) = W^{k,p}$ and $W^{k,2} = H^k$, $k \in \mathbb{N}$, $p \in [1,\infty]$, for classical Sobolev spaces, setting in particular

  $$\|f\|_{H^2}^2 = \|f\|_{L^2}^2 + \|\Delta f\|_{L^2}^2.$$

Further, we use the following inequalities:

- Hölder's inequality on Lorentz spaces [24]. Let $f \in L^{p_1,q_1}$ and $g \in L^{p_2,q_2}$, with $p_1, p_2 \in (0,\infty)$ and $q_1, q_2 \in (0,\infty]$. Then

  $$\|f \cdot g\|_{L^{r,s}} \lesssim_{p_1,p_2,q_1,q_2} \|f\|_{L^{p_1,q_1}} \|g\|_{L^{p_2,q_2}}, \tag{A.1}$$

  with $1/r = 1/p_1 + 1/p_2$ and $1/s = 1/q_1 + 1/q_2$.

- Young's convolution inequality on Lorentz spaces [33, Theorem 2.10.1]. Let $f \in L^{p_1,q_1}$ and $g \in L^{p_2,q_2}$, with $1/p_1 + 1/p_2 > 1$. Then

$$\|f * g\|_{L^{r,s}} \leq 3r\|f\|_{L^{p_1,q_1}}\|g\|_{L^{p_2,q_2}},$$

  with $1/r = 1/p_1 + 1/p_2 - 1$ and $s \in [1,\infty]$ such that $1/q_1 + 1/q_2 \geq 1/s$.
- Hardy's inequality [16]:

$$\||x - \cdot|^{-1}f\|_{L^2} \leq 2\|\nabla f\|_{L^2} \tag{A.2}$$

  for all $f \in H^1$ and $x \in \mathbb{R}^3$.

References

[1] F. Agostini, A. Abedi, Y. Suzuki, E.K.U. Gross, Mixed quantum-classical dynamics on the exact time-dependent potential energy surface: a fresh look at non-adiabatic processes, Mol. Phys. 111 (22–23) (2013) 3625–3664.
[2] A. Anantharaman, E. Cancès, Existence of minimizers for Kohn-Sham models in quantum chemistry, Ann. Inst. Henri Poincaré, Anal. Non Linéaire 26 (6) (2009) 2425–2455.
[3] L. Baudouin, Existence and regularity of the solution of a time dependent Hartree-Fock equation coupled with a classical nuclear dynamics, Rev. Mat. Complut. 18 (2) (2005) 285–314.
[4] C. Bennett, R. Sharpley, The classical interpolation theorems, in: S.-T. Yau, H. Bass, A. Borel, J. Moser (Eds.), Interpolation of Operators, in: Pure and Applied Mathematics, vol. 129, Academic Press, San Diego, 1988, pp. 183–289.
[5] O. Bokanowski, J.L. López, J. Soler, On an exchange interaction model for quantum transport: the Schrödinger-Poisson-Slater system, Math. Models Methods Appl. Sci. 13 (10) (2003) 1397–1412.
[6] A. Bove, G. Da Prato, G. Fano, An existence proof for the Hartree-Fock time-dependent problem with bounded two-body interaction, Commun. Math. Phys. 37 (3) (1974) 183–191.
[7] A. Bove, G. Da Prato, G. Fano, On the Hartree-Fock time-dependent problem, Commun. Math. Phys. 49 (1) (1976) 25–33.
[8] E. Cancès, C. Le Bris, On the time-dependent Hartree-Fock equations coupled with a classical nuclear dynamics, Math. Models Methods Appl. Sci. 9 (7) (1999) 963–990.
[9] F. Castella, L^2 solutions to the Schrödinger-Poisson system: existence, uniqueness, time behaviour, and smoothing effects, Math. Models Methods Appl. Sci. 7 (8) (1997) 1051–1083.
[10] I. Catto, J. Dolbeault, O. Sánchez, J. Soler, Existence of steady states for the Maxwell-Schrödinger-Poisson system: exploring the applicability of the concentration-compactness principle, Math. Models Methods Appl. Sci. 23 (10) (2013) 1915–1938.
[11] T. Cazenave, Semilinear Schrödinger Equations, Courant Lecture Notes, vol. 10, American Mathematical Society, Providence, 2003.
[12] T. Cazenave, A. Haraux, An Introduction to Semilinear Evolution Equations, Oxford University Press, New York, 1998.
[13] J.M. Chadam, R.T. Glassey, Global existence of solutions to the Cauchy problem for time-dependent Hartree equations, J. Math. Phys. 16 (1122) (1975) 1122–1130.
[14] A.J. Cohen, P. Mori-Sánchez, W. Yang, Challenges for density functional theory, Chem. Rev. 112 (2012) 289–320.
[15] R. Crespo-Otero, M. Barbatti, Recent advances and perspectives on nonadiabatic mixed quantum-classical dynamics, Chem. Rev. 118 (15) (2018) 7026–7068.
[16] G.H. Hardy, J.E. Littlewood, G. Pólya, Inequalities, Cambridge Mathematical Library, Cambridge University Press, Cambridge, 1952.
[17] P. Hohenberg, W. Konn, Inhomogeneous electron gas, Phys. Rev. 136 (3B) (1964) 864–871.
[18] J.W. Jerome, Time dependent closed quantum systems: nonlinear Kohn-Sham potential operators and weak solutions, J. Math. Anal. Appl. 429 (2015) 995–1006.
[19] W. Kohn, L.J. Sham, Self-consistent equations including exchange and correlation effects, Phys. Rev. 140 (4A) (1965) A1133–A1138.
[20] C. Le Bris, P.L. Lions, From atoms to crystals: a mathematical journey, Bull. Am. Math. Soc. (N.S.) 42 (3) (2005) 291–363.
[21] E.H. Lieb, Density functionals for Coulomb systems, Int. J. Quant. Chem. 24 (1983) 243–277.
[22] N.J. Mauser, The Schrödinger-Poisson-Xα equation, Appl. Math. Lett. 14 (2001) 759–763.
[23] A.S. Sanz, S. Miret-Artés, C. Meier, J.A. Beswick, I. Burghardt, K.B. Møller, K.H. Hughes, Quantum trajectory methods, in: D.A. Micha, I. Burghardt (Eds.), Quantum Dynamics of Complex Molecular Systems, in: Springer Series in Chemical Physics, vol. 83, Springer-Verlag, Berlin, 2007, pp. 343–422.
[24] R. O'Neil, Convolution operators and $L(p,q)$ spaces, Duke Math. J. 10 (1) (1963) 129–142.
[25] R.G. Parr, W. Yang, Density-Functional Theory of Atoms and Molecules, Oxford University Press, New York, 1989.
[26] J.P. Perdew, S. Kurth, Density functionals for non-relativistic Coulomb systems in the new century, in: M. Marques, C. Fiolhais, F. Nogueira (Eds.), A Primer in Density Functional Theory, in: Lecture Notes in Physics, vol. 620, Springer-Verlag, Berlin, 2003, pp. 1–55.
[27] J.P. Perdew, A. Zunger, Self-interaction correction to density-functional approximations for many-electron systems, Phys. Rev. B 23 (10) (1981) 5048–5079.

[28] M. Sprengel, G. Ciaramella, A. Borzì, A theoretical investigation of time-dependent Kohn-Sham equations: new proofs, Appl. Anal. 100 (10) (2019) 2254–2273.

[29] J.C. Tully, Mixed quantum-classical dynamics, Faraday Discuss. 110 (1998) 407–419.

[30] C.A. Ullrich, Time-Dependent Density-Functional Theory. Concepts and Applications, Oxford University Press, New York, 2012.

[31] K. Yajima, Existence of solutions for Schrödinger evolution equations, Commun. Math. Phys. 110 (3) (1987) 415–426.

[32] S. Zagatti, The Cauchy problem for Hartree-Fock time-dependent equations, Ann. Inst. Henri Poincaré 56 (4) (1992) 357–374.

[33] W.P. Ziemer, Weakly Differentiable Functions. Sobolev Spaces and Functions of Bounded Variation, Springer-Verlag, Berlin, 1989.