EFFECTIVE MAXWELL EQUATIONS FROM TIME-DEPENDENT DENSITY FUNCTIONAL THEORY

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Abstract. The behavior of interacting electrons in a perfect crystal under macroscopic external electric and magnetic fields is studied. Effective Maxwell equations for the macroscopic electric and magnetic fields are derived starting from time-dependent density functional theory. Effective permittivity and permeability coefficients are obtained.

1. Introduction

This paper is a continuation of our study on the macroscopic behavior of interacting electrons in a crystal. In the previous paper [8], we studied the Bloch dynamics of a single electron in a crystal and introduced the Bloch-Wigner transform for studying the semi-classical limit of Schrödinger equation. We also gave a simplified derivation of the Berry curvature term in the effective dynamics. In this paper, we study the collective behavior of the interacting electrons in an insulating crystal under applied electric and magnetic fields. We derive the effective Maxwell equations in this case using systematic asymptotics. In particular, we obtain the effective permittivity and permeability coefficients for these materials.

From a macroscopic viewpoint, the behavior of crystals can be characterized as follows:

(1) Mechanically, crystals respond to applied stress by deforming the crystal lattice.
(2) Crystals respond to applied electric and magnetic fields by distorting the charge-spin distribution, or by motion of free electrons. This generates electro-magnetic responses.

The mechanical and electro-magnetic responses can be coupled together, generating piezo-electric, magnetorestrictive and ferro-elastic effect, etc. The main purpose of this series of work is to provide a systematic understanding of these macroscopic phenomena and derivation of the effective macroscopic models from "first principles".

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As the first principle, we choose to work with the density functional theory [13,14,19] instead of the many-body Schrödinger or Dirac equations. This is because that density functional theory has proven to be extremely successful for the kind of issues we are interested in, and is at the present time the only tractable and yet reliable models for electronic matter. Here by density functional theory, we mostly mean Kohn-Sham density functional theory that rely on orbitals, as is done in this paper. But occasionally we also resort to orbital-free density functional theory, such as the Thomas-Fermi type of models, to illustrate some of the issues. We refer to [1, 2, 4, 6, 15–18] for the mathematical works done on density functional theory. Closely related are the works on Hartree or Hartree-Fock models, which have also been used as the starting point for analyzing the behavior of crystals.

When the crystal is elastically deformed, continuum mechanics model can be derived from the Cauchy-Born rule (extended to electronic structures). This was done for the Thomas-Fermi-von Weiszäcker model in [4]. In a series of works by E and Lu [10–12], the Cauchy-Born rule was validated for nonlinear tightbinding models and Kohn-Sham density functional theory. One of the important ingredients in these works is the identification of sharp stability criteria when the model has exchange-correlation energy which might be non-convex. The issue of stability does not occur in Thomas-Fermi-von Weiszäcker, Hartree or reduced Hartree-Fock model, since these models do not include exchange-correlation energy. Further in this direction, E and Lu studied in [9] the continuum limit of the spin-polarized Thomas-Fermi-von Weiszäcker-Dirac model under external macroscopic magnetic fields. Under stability conditions for plasmon and magmon, a micromagnetics energy functional was derived.

One interesting by-product of the work in [11] is an effective model for the macroscopic electric potential as a result of the crystal deformation, which exhibits a coupling between the mechanical and electric responses.

Cances and Lewin studied the reduced Hartree-Fock model for a crystal under a macroscopic external potential and proved that the implied macroscopic potential satisfies an effective Poisson equation. In particular, they established the validity of the well-known Adler-Wiser formula for the permittivity tensor [5].

In this work, we consider the time-dependent Kohn-Sham density functional theory in the presence of external macroscopic electric and magnetic fields. The questions of interest are whether macroscopic Maxwell equations that describe the electromagnetic fields can be derived in the continuum limit from the underlying microscopic theory, and in particular, how to obtain effective permittivity and permeability for materials from electronic structure models. We resolve these issues using asymptotic analysis. To rigorously justify the asymptotic derivation, one needs to identify correct stability conditions for time dependent models. This will be left to future publications.

The paper is organized as follows. In Section 2, we introduce the time-dependent Kohn-Sham density functional theory. Section 3 describes the model setup and presents the main results. The asymptotic derivation is given in Section 4, Section 5 and Section 6. We make conclusive remarks in Section 7.

2. Time-dependent density functional theory

Time dependent density functional theory (TDDFT) [19] is an extension of (static) density functional theory to the dynamics of interacting electrons. In TDDFT, the electron dynamics is governed by N one-electron time dependent Schrödinger equations with effective one-body Hamiltonian depending on electron density and/or electron current density.[1]

The TDDFT model takes the following form in physical units in $\mathbb{R}^3$,

$$i\hbar\frac{\partial\psi_j}{\partial t} = \frac{1}{2m_e}\left(-i\hbar\nabla - \frac{e}{c}(\boldsymbol{A}+\boldsymbol{A}_{\rm ext})\right)^2\psi_j + e(V+V_{\rm ext})\psi_j, \tag{2.1}$$

$$-\Delta\phi = \frac{e}{\epsilon_0}(\rho - m), \tag{2.2}$$

$$\frac{1}{c}\frac{\partial}{\partial t}\left(\frac{1}{c}\frac{\partial}{\partial t}\boldsymbol{A} + \nabla\phi\right) - \Delta\boldsymbol{A} = \frac{e}{c\epsilon_0}\boldsymbol{J}, \tag{2.3}$$

$$\nabla\cdot\boldsymbol{A} = 0, \tag{2.4}$$

$$V(t,\boldsymbol{x}) = \phi(t,\boldsymbol{x}) + \eta(\rho(t,\boldsymbol{x})). \tag{2.5}$$

Here ψ_j, $j=1,\dots,N$, is the one-particle wave function, $\boldsymbol{A}$ is the vector potential and ϕ is the scalar potential generated by electrons. The electric and magnetic fields are given by

$$\boldsymbol{E} = -\nabla\phi - \frac{\partial\boldsymbol{A}}{\partial t}, \qquad \boldsymbol{B} = \nabla\times\boldsymbol{A}.$$

The system is invariant under the gauge transform,

$$\boldsymbol{A}\to\boldsymbol{A}+\nabla\chi, \qquad \phi\to\phi-\frac{\partial\chi}{\partial t},$$

and hence we fix the Coulomb gauge (2.4) in the model. $\boldsymbol{A}_{\rm ext}$ and $V_{\rm ext}$ are the external vector and scalar potentials. The electron number density and electron current density are denoted by ρ and $\boldsymbol{J}$ respectively in the equations, and are given in terms of $\{\psi_j\}_{j=1}^N$ by

$$\rho(t,\boldsymbol{x}) = \sum_{j=1}^N |\psi_j(t,\boldsymbol{x})|^2, \tag{2.6}$$

$$\boldsymbol{J}(t,\boldsymbol{x}) = \frac{\hbar}{m_e}\sum_{j=1}^N \mathfrak{Im}\left(\psi_j^*(t,\boldsymbol{x})\nabla\psi_j(t,\boldsymbol{x})\right) - \frac{e}{m_e c}\rho(t,\boldsymbol{x})\boldsymbol{A}(t,\boldsymbol{x}). \tag{2.7}$$

[1]When the effective Hamiltonian depends on electron current density, the model is usually called time dependent current density functional theory (TDCDFT) [20,21] in physics literature, although we still use the name of time dependent density functional theory in this paper.

The function $m(\boldsymbol{x})$ is the background charge density contributed by the nuclei. We assume that the nuclei are fixed so that $m(\boldsymbol{x})$ is independent of time. In (2.1), we have the physical constants electron mass m_e, electron charge e, Planck constant $\hbar$, dielectric constant in vacuum ϵ_0 and speed of light in vacuum c.

The electric and magnetic fields are given by vector and scalar potentials (in the Coulomb gauge),

$$E = -\nabla\phi - \frac{1}{c}\frac{\partial}{\partial t}\boldsymbol{A}; \tag{2.8}$$

$$B = \nabla \times \boldsymbol{A}. \tag{2.9}$$

We make some remarks about the model.

(1) The spin is ignored in the above TDDFT model. As a result, only the orbital magnetization is considered, while spin magnetization is not present. The extension to include spin in the model is straightforward though.
(2) We adopt the adiabatic local density approximation [7,22] for the exchange-correlation potential, denoted as η in equation (2.5). This means that the exchange-correlation potential is a function of local electron density only. No exchange-correlation vector potential is included in the model. Generally, the exchange-correlation potential can depend on the local electron current density and the derivatives of electron density and current density. Exchange-correlation vector potential can also be added. The extension to these general models is in principle possible, but will complicate the formulations and derivations in the discussions below.
(3) The model agrees with what physicists commonly use in practical applications (for instance [3]). Of course, whether the model gives a good prediction of the time-evolution of electronic structure depends on the choice of pseudo-potential, the choice of exchange-correlation functional, and sometimes requires additional terms like exchange-correlation vector potential. We will not go into the details of this discussion.

Nondimensionalization and high frequency scaling

We consider the situation when the applied external fields to the system have a much larger characteristic length compared to the atomistic length scale (lattice parameter). For this purpose, we perform nondimensionalization to the set of equations and identify small parameters.

We introduce two sets of units to rescale the system. One is the microscopic unit in which we denote the units of time, length, mass and charge as $[t]$, $[l]$, $[m]$, $[e]$; the other is the macroscopic unit in which we denote the units of time and length as $[T]$, $[L]$. It means that for example the characteristic time scale for macroscopic fields is $[T]$, while that for microscopic fields is $[t]$. We will consider the macroscopic behavior of the system under macroscopic external potentials within the high frequency regime, in other words, the regime

$$[T] \sim [t], \quad [L] \gg [l].$$

The small parameter is identified as $\varepsilon = [l]/[L]$. Physically, the high frequency regime means that we are interested in the dynamics of the electronic structure and the corresponding dynamics of the electromagnetic fields on the time scale that is comparable to the characteristic time scale of the quantum system. At longer time scale, different physical phenomena might occur and is not covered by the results here. In particular, this is different from the scaling used when considering the semi-classical limit.

Using these two sets of units, we can represent all physical constants and quantities in suitable units so that they become nondimensional and have values of order $\mathcal{O}(1)$. For example, Planck constant, vacuum dielectric constant and speed of light can be written as

$$\hbar = 1 \times \frac{[m][l]^2}{[t]}, \quad \epsilon_0 = 1 \times \frac{[e]^2[t]^2}{[m][l]^3}, \quad c = 1 \times \frac{[L]}{[T]}.$$

The temporal and spatial derivatives are rescaled as

$$\frac{\partial}{\partial t} \longrightarrow \frac{1}{[T]}\frac{\partial}{\partial t}, \quad \nabla \longrightarrow \frac{1}{[L]}\nabla.$$

The physical quantities are rewritten as

$$e\boldsymbol{A} = \widetilde{A}\frac{[m][l]^2}{[t]^2}, \quad eV = \widetilde{V}\frac{[m][l]^2}{[t]^2}, \quad \rho = \widetilde{\rho}\frac{1}{[L]^3}, \quad \boldsymbol{J} = \widetilde{\boldsymbol{J}}\frac{[l]^2}{[t][L]^4},$$

where $\widetilde{A}$, $\widetilde{V}$, $\widetilde{\rho}$, $\widetilde{\boldsymbol{J}}$ are nondimensional quantities.

Substituting all the above into the system (2.1)-(2.5) produces the nondimensionalized TDDFT equations (the tildes are dropped for simplicity),

$$i\frac{\partial \psi_j}{\partial t} = \frac{1}{2}\left(-i\varepsilon\nabla - \varepsilon(\boldsymbol{A} + \boldsymbol{A}_{\mathrm{ext}})\right)^2 \psi_j + (V + V_{\mathrm{ext}})\psi_j, \tag{2.10}$$

$$-\Delta\phi = \varepsilon(\rho - m), \tag{2.11}$$

$$\frac{\partial^2}{\partial t^2}\boldsymbol{A} - \Delta\boldsymbol{A} + \frac{\partial}{\partial t}\nabla\phi = \varepsilon^2\boldsymbol{J}, \tag{2.12}$$

$$\nabla \cdot \boldsymbol{A} = 0, \tag{2.13}$$

$$V(t,\boldsymbol{x}) = \phi(t,\boldsymbol{x}) + \eta(\varepsilon^3\rho(t,\boldsymbol{x})). \tag{2.14}$$

The density and current are given by

$$\rho(t,\boldsymbol{x}) = \sum_{j=1}^{N} |\psi_j(t,\boldsymbol{x})|^2, \tag{2.15}$$

$$\boldsymbol{J}(t,\boldsymbol{x}) = \varepsilon\sum_{j=1}^{N} \mathfrak{Im}\left(\psi_j^*(t,\boldsymbol{x})\nabla\psi_j(t,\boldsymbol{x})\right) - \varepsilon\rho(t,\boldsymbol{x})\boldsymbol{A}(t,\boldsymbol{x}). \tag{2.16}$$

3. The effective Maxwell equations in crystal

3.1. **Unperturbed system.** Let $\mathbb{L}$ be a lattice with unit cell Γ. Denote the reciprocal lattice as $\mathbb{L}^*$ and the reciprocal unit cell as Γ^*. We consider system as a crystal $\varepsilon\mathbb{L}$, so that ε is the lattice constant (the micro length scale used in the non-dimensionalization). Therefore, the charge background is given by

$$m^\varepsilon(\boldsymbol{x}) = \varepsilon^{-3} m_0(\boldsymbol{x}/\varepsilon), \tag{3.1}$$

where m_0 is Γ-periodic. Note that the factor ε^{-3} comes from rescaling so that the total background charge in one unit cell is the constant Z independent of ε, *i.e.*

$$\int_{\varepsilon\Gamma} m^\varepsilon(\boldsymbol{x})\,\mathrm{d}\boldsymbol{x} = Z. \tag{3.2}$$

We introduce the following notations for cell average in physical and reciprocal spaces

$$\langle f(\boldsymbol{z})\rangle_{\boldsymbol{z}} = \int_\Gamma f(\boldsymbol{z})\,\mathrm{d}\boldsymbol{z}, \qquad \fint_{\Gamma^*} g(\boldsymbol{k})\,\mathrm{d}\boldsymbol{k} = \frac{1}{|\Gamma^*|}\int_{\Gamma^*} g(\boldsymbol{k})\,\mathrm{d}\boldsymbol{k}.$$

When there are no external applied potentials ($V_{\text{ext}} = 0$, $\boldsymbol{A}_{\text{ext}} = 0$), TDDFT system can be written as

$$i\frac{\partial \psi_j^\varepsilon}{\partial t} = \tfrac{1}{2}\bigl(-i\varepsilon\nabla - \varepsilon\boldsymbol{A}^\varepsilon\bigr)^2\psi_j^\varepsilon + V^\varepsilon\psi_j^\varepsilon, \tag{3.3}$$

$$-\Delta\phi^\varepsilon = \varepsilon\bigl(\rho^\varepsilon(t,\boldsymbol{x}) - m^\varepsilon(\boldsymbol{x})\bigr), \tag{3.4}$$

$$\frac{\partial^2}{\partial t^2}\boldsymbol{A}^\varepsilon - \Delta\boldsymbol{A}^\varepsilon + \frac{\partial}{\partial t}(\nabla\phi^\varepsilon) = \varepsilon^2\boldsymbol{J}^\varepsilon, \quad \nabla\cdot\boldsymbol{A}^\varepsilon = 0, \tag{3.5}$$

$$V^\varepsilon(t,\boldsymbol{x}) = \phi^\varepsilon(t,\boldsymbol{x}) + \eta(\varepsilon^3\rho^\varepsilon(t,\boldsymbol{x})). \tag{3.6}$$

We assume that there exists a ground state for the unperturbed system, with density having the lattice periodicity

$$\rho^\varepsilon(\boldsymbol{x}) = \varepsilon^{-3}\rho_{\text{gs}}(\boldsymbol{x}/\varepsilon), \tag{3.7}$$

where ρ_{gs} Γ-periodic. Absence of external perturbation implies that the system will stay at the ground state with no electronic current and hence no induced vector potential,

$$\boldsymbol{J}^\varepsilon(t,\boldsymbol{x}) = 0, \quad \boldsymbol{A}^\varepsilon(t,\boldsymbol{x}) = 0.$$

The evolution equations are then simplified as

$$i\frac{\partial\psi_j}{\partial t} = -\frac{\varepsilon^2}{2}\Delta\psi_j + V^\varepsilon(\boldsymbol{x})\psi_j, \tag{3.8}$$

$$V^\varepsilon = \phi^\varepsilon(\boldsymbol{x}) + \eta(\rho_{\text{gs}}(\boldsymbol{x}/\varepsilon)), \tag{3.9}$$

$$-\Delta\phi^\varepsilon = \varepsilon^{-2}(\rho_{\text{gs}}(\boldsymbol{x}/\varepsilon) - m_0(\boldsymbol{x}/\varepsilon)). \tag{3.10}$$

Note that the potential is independent of time if there is no external perturbation. It is easy to see that the potential is $\varepsilon\Gamma$-periodic. We denote the potential corresponding to the ground state as $V^\varepsilon(\boldsymbol{x}) = v_0(\boldsymbol{x}/\varepsilon) = v_{\text{gs}}(\boldsymbol{x}/\varepsilon)$ where v_{gs} is Γ-periodic.

The Hamiltonian operator for the ground state is independent of time, given by

$$H_0^\varepsilon = -\frac{\varepsilon^2}{2}\Delta + v_{\mathrm{gs}}(\boldsymbol{x}/\varepsilon). \tag{3.11}$$

Define the rescaling operator δ_ε as

$$(\delta_\varepsilon f)(\boldsymbol{x}) = \varepsilon^{-3/2} f(\boldsymbol{x}/\varepsilon). \tag{3.12}$$

It is easy to check that δ_ε is a unitary operator. We have

$$H_0 = -\frac{1}{2}\Delta + v_0(\boldsymbol{x}) = \delta_\varepsilon^* H_0^\varepsilon \delta_\varepsilon. \tag{3.13}$$

Since v_0 is Γ-periodic, H_0 is invariant under the translation with respect to the lattice $\mathbb{L}$. The standard Bloch-Floquet theory gives the decomposition of H_0,

$$H_0 = ⨍_{\Gamma^*} H_{0,\boldsymbol{k}} \,\mathrm{d}\boldsymbol{k}, \tag{3.14}$$

where $H_{0,\boldsymbol{k}}$ is an operator defined on $L^2_{\boldsymbol{k}}(\Gamma)$ for each $\boldsymbol{k} \in \Gamma^*$,

$$L^2_{\boldsymbol{k}}(\Gamma) = \{f \in L^2(\Gamma) \mid \tau_{\boldsymbol{R}} f = e^{-i\boldsymbol{R}\cdot\boldsymbol{k}} f,\ \forall \boldsymbol{R} \in \mathbb{L}\}.$$

Here $\tau_{\boldsymbol{R}}$ is the translation operator, *i.e.* $\tau_{\boldsymbol{R}} f(\boldsymbol{x}) = f(\boldsymbol{x} + \boldsymbol{R})$. The operator $H_{0,\boldsymbol{k}}$ has the spectral representation

$$H_{0,\boldsymbol{k}} = \sum_n E_n(\boldsymbol{k}) |\psi_{n,\boldsymbol{k}}\rangle\langle\psi_{n,\boldsymbol{k}}|, \tag{3.15}$$

where $E_n(\boldsymbol{k})$ is the n-th eigenvalue of $H_{0,\boldsymbol{k}}$, and $\psi_{n,\boldsymbol{k}}$ is the corresponding eigenfunction (named as Bloch wave in literature) with

$$u_{n,\boldsymbol{k}}(\boldsymbol{x}) = e^{-i\boldsymbol{k}\boldsymbol{x}} \psi_{n,\boldsymbol{k}}(\boldsymbol{x})$$

being Γ-periodic. Moreover, the spectrum $\mathrm{spec}(H_0)$ has the band structure,

$$\mathrm{spec}(H_0) = \bigcup_n \bigcup_{\boldsymbol{k}\in\Gamma^*} E_n(\boldsymbol{k}).$$

Denote the spectrum for the first Z bands by σ_Z,

$$\sigma_Z = \bigcup_{n=1}^{Z} \bigcup_{\boldsymbol{k}\in\Gamma^*} E_n(\boldsymbol{k}), \tag{3.16}$$

where $E_n(\boldsymbol{k})$ is the n-th eigenvalue of H_0.

We assume that the ground state satisfies the gap condition,

$$\mathrm{dist}(\sigma_Z, \mathrm{spec}(H_0)) \backslash \sigma_Z) = E_g. \tag{3.17}$$

In physical terminology, the system is called a band insulator with band gap E_g.

For convenience, we use the bra and ket notations

$$\langle f(\boldsymbol{\zeta}) | \mathcal{K} | g(\boldsymbol{\zeta}) \rangle_{L^2(\Gamma)} = \int_\Gamma f^*(\boldsymbol{\zeta}) \mathcal{K} g(\boldsymbol{\zeta}) \,\mathrm{d}\boldsymbol{\zeta},$$

where $\mathcal{K} : L^2(\Gamma) \to L^2(\Gamma)$ is a linear operator.

3.2. **Macroscopic perturbation.** We are interested in the dynamics of the electronic structure in the presence of the external potentials $\boldsymbol{A}_{\text{ext}}(t,\boldsymbol{x})$ and $V_{\text{ext}}(t,\boldsymbol{x})$. We assume that $\boldsymbol{A}_{\text{ext}}$ and V_{ext} are smooth functions in both t and $\boldsymbol{x}$ and periodic in space in the domain Γ. Hence, the characteristic length scales of external applied fields are $\mathcal{O}(1)$, while the lattice constant is $\mathcal{O}(\varepsilon)$. We consider the continuum limit $\varepsilon \to 0$; the disparity of the space scales leads to macroscopic Maxwell equations.

We consider the following system with periodic conditions on Γ,

$$i\frac{\partial \psi_j^\varepsilon}{\partial t} = H^\varepsilon \psi_j^\varepsilon, \tag{3.18}$$

$$-\Delta\phi^\varepsilon = \varepsilon\big(\rho^\varepsilon(t,\boldsymbol{x}) - m_0(\boldsymbol{x}/\varepsilon)\big), \tag{3.19}$$

$$\frac{\partial^2}{\partial t^2}\boldsymbol{A}^\varepsilon - \Delta\boldsymbol{A}^\varepsilon + \frac{\partial}{\partial t}\left(\nabla\phi^\varepsilon\right) = \varepsilon^2\boldsymbol{J}^\varepsilon, \tag{3.20}$$

$$\nabla\cdot\boldsymbol{A}^\varepsilon = 0, \tag{3.21}$$

$$V^\varepsilon(t,\boldsymbol{x}) = \phi^\varepsilon(t,\boldsymbol{x}) + \eta(\varepsilon^3\rho^\varepsilon(t,\boldsymbol{x})), \tag{3.22}$$

where the Hamiltonian operator H^ε is given by

$$H^\varepsilon = \frac{1}{2}\big(-i\varepsilon\nabla - \varepsilon(\boldsymbol{A}^\varepsilon + \boldsymbol{A}_{\text{ext}})\big)^2 + V^\varepsilon + V_{\text{ext}}.$$

We have used the superscript ε to make explicit the dependence on the small parameter. The density and current is then given by

$$\rho^\varepsilon = \sum_{k=1}^{Z/\varepsilon^3}\left|\psi_j^\varepsilon\right|^2, \quad \boldsymbol{J}^\varepsilon = \varepsilon\sum_{k=1}^{Z/\varepsilon^3}\mathfrak{Im}\,\big((\psi_j^\varepsilon)^*\nabla\psi_j^\varepsilon\big) - \varepsilon\boldsymbol{A}^\varepsilon\rho^\varepsilon.$$

Here Z is the number of electrons in one unit cell, which equals to the background charge (3.2). We remark that in the domain Γ, since the lattice constant is ε, there are ε^{-3} unit cells in total, and hence $N = Z\varepsilon^{-3}$ electrons under consideration.

3.3. **Main result.** Define the limiting macroscopic potentials as

$$U_0(t,\boldsymbol{x}) = \lim_{\varepsilon\to 0}\big(V^\varepsilon(t,\boldsymbol{x}) + V_{\text{ext}}(t,\boldsymbol{x}) - v_{\text{gs}}(\boldsymbol{x}/\varepsilon)\big), \tag{3.23}$$

$$A_0(t,\boldsymbol{x}) = \lim_{\varepsilon\to 0}\big(A^\varepsilon(t,\boldsymbol{x}) + A_{\text{ext}}(t,\boldsymbol{x})\big); \tag{3.24}$$

and the corresponding electric and magnetic fields

$$\boldsymbol{E}(t,\boldsymbol{x}) = -\nabla_{\boldsymbol{x}}U_0(t,\boldsymbol{x}) - \frac{\partial}{\partial t}\boldsymbol{A}_0(t,\boldsymbol{x}), \tag{3.25}$$

$$\boldsymbol{B}(t,\boldsymbol{x}) = \nabla_{\boldsymbol{x}}\times\boldsymbol{A}_0(t,\boldsymbol{x}). \tag{3.26}$$

Define the electric field in frequency space,

$$\widehat{\boldsymbol{E}}(\omega,\boldsymbol{x}) = \int_0^\infty e^{i\omega t}\boldsymbol{E}(t,\boldsymbol{x})\,\mathrm{d}t,$$

and similarly for $\widehat{\boldsymbol{B}}$, $\widehat{U_0}$ and $\widehat{\boldsymbol{A}_0}$. We have

$$\widehat{\boldsymbol{E}}(\omega,\boldsymbol{x}) = -\nabla_{\boldsymbol{x}}\widehat{U_0}(\omega,\boldsymbol{x}) + i\omega\widehat{\boldsymbol{A}_0}(\omega,\boldsymbol{x}), \tag{3.27}$$

$$\widehat{\boldsymbol{B}}(\omega,\boldsymbol{x}) = \nabla_{\boldsymbol{x}} \times \widehat{\boldsymbol{A}_0}(\omega,\boldsymbol{x}). \tag{3.28}$$

We will show that TDDFT system gives arise to the effective Maxwell system as

$$\nabla_{\boldsymbol{x}} \cdot \big(\mathcal{E}(\omega)\widehat{\boldsymbol{E}}(\omega,\boldsymbol{x})\big) = \widehat{\rho_{\rm ext}}(\omega,\boldsymbol{x}), \tag{3.29}$$

$$\nabla_{\boldsymbol{x}} \cdot \widehat{\boldsymbol{B}}(\omega,\boldsymbol{x}) = 0, \tag{3.30}$$

$$\nabla_{\boldsymbol{x}} \times \widehat{\boldsymbol{E}}(\omega,\boldsymbol{x}) = i\omega\widehat{\boldsymbol{B}}(\omega,\boldsymbol{x}), \tag{3.31}$$

$$\nabla_{\boldsymbol{x}} \times \widehat{\boldsymbol{B}}(\omega,\boldsymbol{x}) = -i\omega\mathcal{E}(\omega)\widehat{\boldsymbol{E}}(\omega,\boldsymbol{x}) + \widehat{\boldsymbol{J}_{\rm ext}}(\omega,\boldsymbol{x}), \tag{3.32}$$

where

$$\begin{aligned}
\widehat{\rho}_{\rm ext}(\omega,\boldsymbol{x}) &= \int_0^\infty e^{i\omega t}\rho_{\rm ext}(t,\boldsymbol{x})\,\mathrm{d}t,\\
\widehat{\boldsymbol{J}}_{\rm ext}(\omega,\boldsymbol{x}) &= \int_0^\infty e^{i\omega t}\boldsymbol{J}_{\rm ext}(t,\boldsymbol{x})\,\mathrm{d}t,
\end{aligned}$$

with $\rho_{\rm ext}$ and $\boldsymbol{J}_{\rm ext}$ given by

$$\begin{aligned}
\rho_{\rm ext}(t,\boldsymbol{x}) &= -\Delta_{\boldsymbol{x}} V_{\rm ext}(t,\boldsymbol{x}),\\
\boldsymbol{J}_{\rm ext}(t,\boldsymbol{x}) &= \frac{\partial^2}{\partial t^2}\boldsymbol{A}_{\rm ext} - \Delta_{\boldsymbol{x}}\boldsymbol{A}_{\rm ext} + \frac{\partial}{\partial t}\left(\nabla_{\boldsymbol{x}} V_{\rm ext}\right).
\end{aligned}$$

The system (3.29)-(3.32) are (nonlocal) Maxwell equations with dynamic dielectric permittivity matrix $\mathcal{E}_{\alpha\beta} = \delta_{\alpha\beta} + A_{\alpha\beta}$ given by

$$\begin{aligned}
A_{\alpha\beta}(\omega) = &\\
&\sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega+\omega_{mn}(\boldsymbol{k})}\overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)}\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\,\mathrm{d}\boldsymbol{k}\\
&-\sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega-\omega_{mn}(\boldsymbol{k})}\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)}\,\mathrm{d}\boldsymbol{k}\\
&-\frac{2i}{\omega}\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\fint_{\Gamma^*}\overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle}\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{m,\boldsymbol{k}}\rangle\,\mathrm{d}\boldsymbol{k}\\
&-\left\langle \widehat{f}^*_\alpha(\omega,\boldsymbol{z})\mathcal{V}\left(\mathcal{I}-\widehat{\chi}_\omega\mathcal{V}\right)^{-1}\widehat{f}_\beta(\omega,\boldsymbol{z})\right\rangle_{\boldsymbol{z}}.
\end{aligned}$$

Here the potential operator $\mathcal{V}$ is the linearized effective potential operator at the equilibrium density ρ_0:

$$\begin{aligned}
&(\mathcal{V}f)(\boldsymbol{z}) = \phi(\boldsymbol{z}) + \eta'(\rho_0(\boldsymbol{z}))f(\boldsymbol{z}),\\
&-\Delta_{\boldsymbol{z}}\phi(\boldsymbol{z}) = f(\boldsymbol{z}), \quad \langle\phi\rangle = 0.
\end{aligned}$$

The operator $\widehat{\chi}_\omega$ and the function $\widehat{\boldsymbol{f}}$ are defined as

$$\begin{aligned}\widehat{\chi}_\omega v = &- \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega+\omega_{mn}(\boldsymbol{k})} u_{n,\boldsymbol{k}} u_{m,\boldsymbol{k}}^* \langle u_{n,\boldsymbol{k}} | v | u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \,\mathrm{d}\boldsymbol{k} \\ &+ \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega-\omega_{mn}(\boldsymbol{k})} u_{n,\boldsymbol{k}}^* u_{m,\boldsymbol{k}} \overline{\langle u_{n,\boldsymbol{k}} | v | u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)} \,\mathrm{d}\boldsymbol{k}, \\ \widehat{\boldsymbol{f}}(\omega) = &- \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega+\omega_{mn}(\boldsymbol{k})} u_{n,\boldsymbol{k}} u_{m,\boldsymbol{k}}^* \langle u_{n,\boldsymbol{k}} | i\nabla_{\boldsymbol{k}} | u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \,\mathrm{d}\boldsymbol{k} \\ &+ \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega-\omega_{mn}(\boldsymbol{k})} u_{n,\boldsymbol{k}}^* u_{m,\boldsymbol{k}} \overline{\langle u_{n,\boldsymbol{k}} | i\nabla_{\boldsymbol{k}} | u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)} \,\mathrm{d}\boldsymbol{k}.\end{aligned}$$

Remark that the dynamic permittivity matrix $\mathcal{E}$ is completely determined by the linear response of the unperturbed electronic structure.

The Maxwell equations (3.29)-(3.32) are nonlocal in time, since the permittivity $\mathcal{E}$ depends on ω. While the system is local in space due to the limit $\varepsilon \to 0$, the nonlocality in time is natural since there is no scale separation in time.

We also note that while we obtain a nontrivial effective permittivity, the effective permeability in the equation equals to 1, the same value as in the vacuum. Physically, this is consistent with the situation of semiconductors or insulators under consideration here.

4. Asymptotic analysis of the Schrödinger-Maxwell equations

To derive the effective Maxwell equation, let us take the following ansatz for the system (3.18)-(3.22),

$$\rho^\varepsilon(t,\boldsymbol{x}) = \varepsilon^{-3}\rho_0(\boldsymbol{x}/\varepsilon) + \varepsilon^{-2}\rho_1(t,\boldsymbol{x},\boldsymbol{x}/\varepsilon) + \varepsilon^{-1}\rho_2(t,\boldsymbol{x},\boldsymbol{x}/\varepsilon) + \cdots, \tag{4.1}$$

$$\boldsymbol{J}^\varepsilon(t,\boldsymbol{x}) = \varepsilon^{-3}\boldsymbol{J}_0(\boldsymbol{x}/\varepsilon) + \varepsilon^{-2}\boldsymbol{J}_1(t,\boldsymbol{x},\boldsymbol{x}/\varepsilon) + \cdots, \tag{4.2}$$

$$\boldsymbol{A}^\varepsilon(t,\boldsymbol{x}) + \boldsymbol{A}_{\mathrm{ext}}(t,\boldsymbol{x}) = \boldsymbol{A}_0(t,\boldsymbol{x},\boldsymbol{x}/\varepsilon) + \varepsilon\boldsymbol{A}_1(t,\boldsymbol{x},\boldsymbol{x}/\varepsilon) + \cdots, \tag{4.3}$$

$$\begin{aligned}\phi^\varepsilon(t,\boldsymbol{x}) + V_{\mathrm{ext}}(t,\boldsymbol{x}) = \phi_0(t,\boldsymbol{x},\boldsymbol{x}/\varepsilon) &+ \varepsilon\phi_1(t,\boldsymbol{x},\boldsymbol{x}/\varepsilon) \\ &+ \varepsilon^2\phi_2(t,\boldsymbol{x},\boldsymbol{x}/\varepsilon) + \cdots,\end{aligned} \tag{4.4}$$

where the higher order terms are omitted. We also assume that the dependence on the fast variable $\boldsymbol{z} = \boldsymbol{x}/\varepsilon$ is periodic for all these functions.

The main strategy of asymptotic analysis is as follows. We first apply a two-scale expansion on the Maxwell equations (3.19)-(3.20), which produces the asymptotics of Hamiltonian; then by Dyson series we obtain the asymptotics of density and current; the effective equations in time domain are derived by taking the $\boldsymbol{z}$-average on the second order perturbation equations of the Coulomb potential and vector potential. The asymptotics is somewhat nontrivial. The Coulomb interaction makes the leading order potential dependent on the macroscopic average of the third order density. To close the asymptotics, one has to show that the macroscopic average of the third order density only depends on the leading order potential, but not on higher order terms of the potential. This amounts to establishing the local neutrality of the system, which will be explained in detail below. Finally Fourier transform gives the effective Maxwell equation in frequency domain. Note that we have assumed that the leading order density and current only depend on the fast variable $\boldsymbol{x}/\varepsilon$. This will be justified by the asymptotics.

4.1. **Asymptotics of the Hamiltonian.** For the Coulomb potential, substituting the ansatz (4.1) and (4.4) in (3.19) and organizing the results in orders, one has

$$-\Delta_{\boldsymbol{z}}\phi_0 = \rho_0 - m_0, \tag{4.5}$$

$$-\Delta_{\boldsymbol{z}}\phi_1 - 2\nabla_{\boldsymbol{x}}\cdot\nabla_{\boldsymbol{z}}\phi_0 = \rho_1, \tag{4.6}$$

$$-\Delta_{\boldsymbol{z}}\phi_2 - 2\nabla_{\boldsymbol{x}}\cdot\nabla_{\boldsymbol{z}}\phi_1 - \Delta_{\boldsymbol{x}}\phi_0 = \rho_2 + \rho_{\mathrm{ext}}. \tag{4.7}$$

Recall that $\rho_{\mathrm{ext}}(t,\boldsymbol{x}) = -\Delta_{\boldsymbol{x}}V_{\mathrm{ext}}(t,\boldsymbol{x})$.

For the exchange-correlation potential, Taylor expansion yields

$$\begin{aligned}\eta(\varepsilon^3\rho^\varepsilon) &= \eta(\rho_0) + \varepsilon\eta'(\rho_0)\rho_1 + \tfrac{1}{2}\varepsilon^2\eta''(\rho_0)\rho_1^2 + \varepsilon^2\eta'(\rho_0)\rho_2 + \cdots\\ &= \eta_0 + \varepsilon\eta_1 + \varepsilon^2\eta_2 + \cdots\ ,\end{aligned} \tag{4.8}$$

where the last equality gives the definition of $\eta_i(t,\boldsymbol{x},\boldsymbol{z})$,

$$\begin{aligned}&\eta_0(\boldsymbol{z}) = \eta(\rho_0(\boldsymbol{z})),\\ &\eta_1(t,\boldsymbol{x},\boldsymbol{z}) = \eta'(\rho_0(\boldsymbol{z}))\rho_1(t,\boldsymbol{x},\boldsymbol{z}),\\ &\eta_2(t,\boldsymbol{x},\boldsymbol{z}) = \tfrac{1}{2}\eta''(\rho_0(\boldsymbol{z}))\rho_1(t,\boldsymbol{x},\boldsymbol{z})^2 + \eta'(\rho_0(\boldsymbol{z}))\rho_2(t,\boldsymbol{x},\boldsymbol{z}),\end{aligned}$$

and similarly for higher order terms, which we omitted in the expression.

Therefore, the total potential V^ε can be written as

$$\begin{aligned}V^\varepsilon &= \phi + V_{\mathrm{ext}} + \eta\\ &= (\phi_0+\eta_0) + \varepsilon(\phi_1+\eta_1) + \varepsilon^2(\phi_2+\eta_2) + \cdots\\ &= V_0 + \varepsilon V_1 + \varepsilon^2 V_2 + \cdots\ .\end{aligned} \tag{4.9}$$

Similarly, we write down the equations for the vector potential using ansatz (4.2) and (4.3):

$$-\Delta_{\boldsymbol{z}}\boldsymbol{A}_0 = 0, \tag{4.10}$$

$$-\Delta_{\boldsymbol{z}}\boldsymbol{A}_1 - 2\nabla_{\boldsymbol{x}}\cdot\nabla_{\boldsymbol{z}}\boldsymbol{A}_0 + \frac{\partial}{\partial t}\big(\nabla_{\boldsymbol{z}}V_0\big) = \boldsymbol{J}_0, \tag{4.11}$$

$$\begin{aligned}&\frac{\partial^2}{\partial t^2}\boldsymbol{A}_0 - \Delta_{\boldsymbol{x}}\boldsymbol{A}_0 - \nabla_{\boldsymbol{z}}\boldsymbol{A}_2 - 2\nabla_{\boldsymbol{x}}\cdot\nabla_{\boldsymbol{z}}\boldsymbol{A}_1\\ &\qquad\qquad + \frac{\partial}{\partial t}\big(\nabla_{\boldsymbol{z}}V_1 + \nabla_{\boldsymbol{x}}V_0\big) = \boldsymbol{J}_1 + \boldsymbol{J}_{\mathrm{ext}}.\end{aligned} \tag{4.12}$$

Recall that $\boldsymbol{J}_{\mathrm{ext}}(t,\boldsymbol{x}) = \frac{\partial^2}{\partial t^2}\boldsymbol{A}_{\mathrm{ext}} - \Delta_{\boldsymbol{x}}\boldsymbol{A}_{\mathrm{ext}} + \frac{\partial}{\partial t}\left(\nabla_{\boldsymbol{x}}V_{\mathrm{ext}}\right)$.

Note that the solvability conditions of (4.5)-(4.6) and (4.11) impose the following constraint on ρ_0, ρ_1 and $\boldsymbol{J}_0$,

$$\langle \rho_0(\boldsymbol{z})\rangle_{\boldsymbol{z}} = \langle m_0(\boldsymbol{z})\rangle_{\boldsymbol{z}}, \quad \langle \rho_1(t,\boldsymbol{x},\boldsymbol{z})\rangle_{\boldsymbol{z}} = 0, \quad \langle \boldsymbol{J}_0(\boldsymbol{z})\rangle_{\boldsymbol{z}} = 0. \tag{4.13}$$

Therefore, the Hamiltonian operator can be written as

$$\begin{aligned} H^\varepsilon(t) = -\tfrac{1}{2}\varepsilon^2\Delta_{\boldsymbol{x}} &+ v_0(\boldsymbol{x}/\varepsilon) + U_0(t,\boldsymbol{x}) + \varepsilon v_1(t,\boldsymbol{x},\boldsymbol{x}/\varepsilon) \\ &+ \varepsilon U_1(t,\boldsymbol{x}) + i\varepsilon^2\boldsymbol{A}_0(t,\boldsymbol{x})\cdot\nabla_{\boldsymbol{x}} + \varepsilon^2 V_2(t,\boldsymbol{x},\boldsymbol{x}/\varepsilon) \\ &+ \varepsilon^2\frac{|\boldsymbol{A}_0(t,\boldsymbol{x})|^2}{2} + i\varepsilon^3\boldsymbol{A}_1(t,\boldsymbol{x},\boldsymbol{x}/\varepsilon)\cdot\nabla_{\boldsymbol{x}} + \cdots \end{aligned} \tag{4.14}$$

where we omit the higher order terms. In (4.14), we define $v_0,\ v_1$ and $U_0,\ U_1$ as the *microscopic* and *macroscopic* components of $V_0,\ V_1$ respectively,

$$\begin{aligned} U_0(t,\boldsymbol{x}) &= \langle V_0(t,\boldsymbol{x},\cdot)\rangle_{\boldsymbol{z}}, \quad v_0(t,\boldsymbol{x},\boldsymbol{z}) = V_0(t,\boldsymbol{x},\boldsymbol{z}) - U_0(t,\boldsymbol{x}); \\ U_1(t,\boldsymbol{x}) &= \langle V_1(t,\boldsymbol{x},\cdot)\rangle_{\boldsymbol{z}}, \quad v_1(t,\boldsymbol{x},\boldsymbol{z}) = V_1(t,\boldsymbol{x},\boldsymbol{z}) - U_1(t,\boldsymbol{x}). \end{aligned} \tag{4.15}$$

4.2. **Asymptotics of the density and current.** The initial state is given by the ground state of the unperturbed system, which implies that the density matrix is given by the projection operator to the occupied spectrum of the ground states in the beginning (with lattice parameter ε),

$$\boldsymbol{\rho}^\varepsilon(0) = \mathcal{P}^\varepsilon. \tag{4.16}$$

Then, the density matrix at time t is given by[2]

$$\boldsymbol{\rho}^\varepsilon(t) = \mathcal{T}\exp\Big(-i\int_0^t H^\varepsilon(\tau)\,\mathrm{d}\tau\Big)\mathcal{P}^\varepsilon\Big(\mathcal{T}\exp\Big(-i\int_0^t H^\varepsilon(\tau)\,\mathrm{d}\tau\Big)\Big)^*, \tag{4.17}$$

where $\mathcal{T}$ is the time ordering operator. Therefore the density is given by the diagonal of the kernel of the operator $\boldsymbol{\rho}^\varepsilon$,

$$\begin{aligned} \rho^\varepsilon(t,\boldsymbol{x}) = \boldsymbol{\rho}^\varepsilon(t,\boldsymbol{x},\boldsymbol{x}) = \mathcal{T}\exp\Big(-i\int_0^t H^\varepsilon(\tau)\,\mathrm{d}\tau\Big) \\ \times \mathcal{P}^\varepsilon\Big(\mathcal{T}\exp\Big(-i\int_0^t H^\varepsilon(\tau)\,\mathrm{d}\tau\Big)\Big)^*(\boldsymbol{x},\boldsymbol{x}), \end{aligned} \tag{4.18}$$

where the right hand side means the diagonal of the kernel associated with the operator. The ground state density is given by $\rho^\varepsilon_{\mathrm{gs}}(\boldsymbol{x}) = \mathcal{P}^\varepsilon(\boldsymbol{x},\boldsymbol{x})$.

We investigate the asymptotic expansion of $\rho^\varepsilon(t,\boldsymbol{x})$ determined by the Hamiltonian (4.14). The equation (4.18) implies, to obtain the density at $(t,\boldsymbol{x})$, we first evolve the system backwards to the initial time, project it onto the ground states of the unperturbed system, then evolve it forwards in time to t at $\boldsymbol{x}$. Since the current time scaling is $\mathcal{O}(1)$, when ε goes to 0, the domain of dependence and domain of influence are also of the scale $\mathcal{O}(\varepsilon)$ for the system in the time evolution. In other words, the density at $(t,\boldsymbol{x})$ only depends on the Hamiltonian of a small

[2]In the language of physics, we are using the Heisenberg picture.

neighborhood $(0,t)\times B(\boldsymbol{x},\mathcal{O}(\varepsilon))$, where $B(\boldsymbol{x},\mathcal{O}(\varepsilon))$ indicates a ball centered at $\boldsymbol{x}$ with the radius $\mathcal{O}(\varepsilon)$.

Accordingly for a fixed point $\boldsymbol{x}\in\Gamma$, we expand the Hamiltonian operator H around $\boldsymbol{x}$. For clarity, we write H as an operator on $L^2_{\boldsymbol{y}}(\Gamma)$,

$$\begin{aligned} H^\varepsilon(t) = -\tfrac{1}{2}\varepsilon^2\Delta_{\boldsymbol{y}} + v_0(\boldsymbol{y}/\varepsilon) + U_0(t,\boldsymbol{y}) + \varepsilon v_1(t,\boldsymbol{y},\boldsymbol{y}/\varepsilon) \\ + \varepsilon U_1(t,\boldsymbol{y}) + i\varepsilon^2\boldsymbol{A}_0(t,\boldsymbol{y})\cdot\nabla_{\boldsymbol{y}} + \varepsilon^2 V_2(t,\boldsymbol{y},\boldsymbol{y}/\varepsilon) \\ + \varepsilon^2\frac{|\boldsymbol{A}_0(t,\boldsymbol{y})|^2}{2} + i\varepsilon^3\boldsymbol{A}_1(t,\boldsymbol{y},\boldsymbol{y}/\varepsilon)\cdot\nabla_{\boldsymbol{y}} + \cdots . \end{aligned}$$

Let $H_0^\varepsilon(t,\boldsymbol{x})$ be the leading order operator

$$H_0^\varepsilon(t,\boldsymbol{x}) = -\tfrac{1}{2}\varepsilon^2\Delta_{\boldsymbol{y}} + v_0(\boldsymbol{y}/\varepsilon) + U_0(t,\boldsymbol{x}). \tag{4.19}$$

Here $H_0^\varepsilon(t,\boldsymbol{x})$ is an operator on $L^2_{\boldsymbol{y}}$ with $(t,\boldsymbol{x})$ as parameters. Similar notations will be used throughout the paper. Denote the difference as $\delta H^\varepsilon(t,\boldsymbol{x}) = H^\varepsilon(t) - H_0^\varepsilon(t,\boldsymbol{x})$,

$$\begin{aligned} \delta H^\varepsilon(t,\boldsymbol{x}) = U_0(t,\boldsymbol{y}) - U_0(t,\boldsymbol{x}) + \varepsilon v_1(t,\boldsymbol{y},\boldsymbol{y}/\varepsilon) + \varepsilon U_1(t,\boldsymbol{y}) \\ + i\varepsilon^2\boldsymbol{A}_0(t,\boldsymbol{y})\cdot\nabla_{\boldsymbol{y}} + \varepsilon^2 V_2(t,\boldsymbol{y},\boldsymbol{y}/\varepsilon) + \varepsilon^2\frac{|\boldsymbol{A}_0(t,\boldsymbol{y})|^2}{2} \\ + i\varepsilon^3\boldsymbol{A}_1(t,\boldsymbol{y},\boldsymbol{y}/\varepsilon)\cdot\nabla_{\boldsymbol{y}} + \cdots . \end{aligned} \tag{4.20}$$

Expand $\delta H^\varepsilon(t,\boldsymbol{x})$ into orders, and the first two orders are

$$\delta H_1^\varepsilon(t,\boldsymbol{x}) = (\boldsymbol{y}-\boldsymbol{x})\cdot\nabla_{\boldsymbol{x}}U_0(t,\boldsymbol{x}) + \varepsilon v_1(t,\boldsymbol{x},\boldsymbol{y}/\varepsilon) + \varepsilon U_1(t,\boldsymbol{x}) + i\varepsilon\boldsymbol{A}_0(t,\boldsymbol{x})\cdot\varepsilon\nabla_{\boldsymbol{y}}, \tag{4.21}$$

and

$$\begin{aligned} \delta H_2^\varepsilon(t,\boldsymbol{x}) =& \tfrac{1}{2}\big((\boldsymbol{y}-\boldsymbol{x})\cdot\nabla_{\boldsymbol{x}}\big)^2 U_0(t,\boldsymbol{x}) + \varepsilon(\boldsymbol{y}-\boldsymbol{x})\cdot\nabla_{\boldsymbol{x}}v_1(t,\boldsymbol{x},\boldsymbol{y}/\varepsilon) \\ &+ \varepsilon(\boldsymbol{y}-\boldsymbol{x})\cdot\nabla_{\boldsymbol{x}}U_1(t,\boldsymbol{x}) + \varepsilon^2 V_2(t,\boldsymbol{x},\boldsymbol{y}/\varepsilon) + \varepsilon^2\frac{|\boldsymbol{A}_0(t,\boldsymbol{x})|^2}{2} \\ &+ i\big(\varepsilon(\boldsymbol{y}-\boldsymbol{x})\cdot\nabla_{\boldsymbol{x}}\boldsymbol{A}_0(t,\boldsymbol{x}) + \varepsilon^2\boldsymbol{A}_1(t,\boldsymbol{x},\boldsymbol{y}/\varepsilon)\big)\cdot\varepsilon\nabla_{\boldsymbol{y}}. \end{aligned} \tag{4.22}$$

Recall that we are approximating the Hamiltonian in a ball around $\boldsymbol{x}$ with the radius $\mathcal{O}(\varepsilon)$, and hence $(\boldsymbol{y}-\boldsymbol{x})$ is treated as $\mathcal{O}(\varepsilon)$ in the above expansion.

We expand the time propagation operator as

$$
\begin{aligned}
\mathcal{T}\exp\Bigl(-i\int_0^t H^\varepsilon(\tau)\,\mathrm{d}\tau\Bigr) =& \mathcal{U}^{\varepsilon,0}_{t,0}(\boldsymbol{x}) \\
& - i\int_0^t \mathcal{T}\exp\Bigl(-i\int_\tau^t H^\varepsilon(s)\,\mathrm{d}s\Bigr)\delta H^\varepsilon(\tau,\boldsymbol{x})\mathcal{U}^{\varepsilon,0}_{\tau,0}(\boldsymbol{x})\,\mathrm{d}\tau \\
=& \mathcal{U}^{\varepsilon,0}_{t,0}(\boldsymbol{x}) - i\int_0^t \mathcal{U}^{\varepsilon,0}_{t,\tau}(\boldsymbol{x})\delta H^\varepsilon(\tau,\boldsymbol{x})\mathcal{U}^{\varepsilon,0}_{\tau,0}(\boldsymbol{x})\,\mathrm{d}\tau \\
& - \int_0^t\int_0^{\tau_2} \mathcal{U}^{\varepsilon,0}_{t,\tau_2}(\boldsymbol{x})\delta H^\varepsilon(\tau_2,\boldsymbol{x})\mathcal{U}^{\varepsilon,0}_{\tau_2,\tau_1}(\boldsymbol{x}) \\
& \qquad \times \delta H^\varepsilon(\tau_1,\boldsymbol{x})\mathcal{U}^{\varepsilon,0}_{\tau_1,0}(\boldsymbol{x})\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2 \\
& + \cdots,
\end{aligned}
$$

where $\mathcal{U}^{\varepsilon,0}_{t_1,t_2}(\boldsymbol{x})$ is the propagation operator corresponding to H^ε_0,

$$
\mathcal{U}^{\varepsilon,0}_{t_1,t_2}(\boldsymbol{x}) = \mathcal{T}\exp\Bigl(-i\int_{t_2}^{t_1} H^\varepsilon_0(\tau,\boldsymbol{x})\,\mathrm{d}\tau\Bigr).
$$

Therefore by (4.21)-(4.22),

$$
\begin{aligned}
\mathcal{T}\exp\Bigl(-i\int_0^t H^\varepsilon(\tau)\Bigr)\,\mathrm{d}\tau =& \mathcal{U}^{\varepsilon,0}_{t,0}(\boldsymbol{x}) - i\int_0^t \mathcal{U}^{\varepsilon,0}_{t,\tau}(\boldsymbol{x})\delta H^\varepsilon_1(\tau,\boldsymbol{x})\mathcal{U}^{\varepsilon,0}_{\tau,0}(\boldsymbol{x})\,\mathrm{d}\tau \\
& - i\int_0^t \mathcal{U}^{\varepsilon,0}_{t,\tau}(\boldsymbol{x})\delta H^\varepsilon_2(\tau,\boldsymbol{x})\mathcal{U}^{\varepsilon,0}_{\tau,0}(\boldsymbol{x})\,\mathrm{d}\tau \\
& - \int_0^t\int_0^{\tau_2} \mathcal{U}^{\varepsilon,0}_{t,\tau_2}(\boldsymbol{x})\delta H^\varepsilon_1(\tau_2,\boldsymbol{x})\mathcal{U}^{\varepsilon,0}_{\tau_2,\tau_1}(\boldsymbol{x}) \\
& \qquad \times \delta H^\varepsilon_1(\tau_1,\boldsymbol{x})\mathcal{U}^{\varepsilon,0}_{\tau_1,0}(\boldsymbol{x})\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2 + \cdots \\
=& \mathcal{U}^{\varepsilon,0}_{t,0}(\boldsymbol{x}) + \varepsilon\mathcal{U}^{\varepsilon,1}_{t,0}(\boldsymbol{x}) + \varepsilon^2\mathcal{U}^{\varepsilon,2}_{t,0}(\boldsymbol{x}) + \cdots,
\end{aligned}
\tag{4.23}
$$

where the last equality defines $\mathcal{U}^{\varepsilon,j}_{t,0}(\boldsymbol{x})$ for $j = 0, 1, 2$. Higher order terms can be written down similarly.

Substituting the expansion (4.23) in (4.18), we obtain to the leading order

$$
\rho^\varepsilon_0(t,\boldsymbol{x}) = \Bigl(\mathcal{U}^{\varepsilon,0}_{t,0}(\boldsymbol{x})\mathcal{P}^\varepsilon\mathcal{U}^{\varepsilon,0}_{0,t}(\boldsymbol{x})\Bigr)(\boldsymbol{x},\boldsymbol{x}),
$$

and also to the higher orders,

$$
\begin{aligned}
\rho^\varepsilon_1(t,\boldsymbol{x}) &= \Bigl(\mathcal{U}^{\varepsilon,0}_{t,0}(\boldsymbol{x})\mathcal{P}^\varepsilon\mathcal{U}^{1,\varepsilon}_{0,t}(\boldsymbol{x})\Bigr)(\boldsymbol{x},\boldsymbol{x}) + \Bigl(\mathcal{U}^{\varepsilon,1}_{t,0}(\boldsymbol{x})\mathcal{P}^\varepsilon\mathcal{U}^{\varepsilon,0}_{0,t}(\boldsymbol{x})\Bigr)(\boldsymbol{x},\boldsymbol{x}), \\
\rho^\varepsilon_2(t,\boldsymbol{x}) &= \Bigl(\mathcal{U}^{\varepsilon,0}_{t,0}(\boldsymbol{x})\mathcal{P}^\varepsilon\mathcal{U}^{2,\varepsilon}_{0,t}(\boldsymbol{x})\Bigr)(\boldsymbol{x},\boldsymbol{x}) + \Bigl(\mathcal{U}^{\varepsilon,1}_{t,0}(\boldsymbol{x})\mathcal{P}^\varepsilon\mathcal{U}^{\varepsilon,1}_{0,t}(\boldsymbol{x})\Bigr)(\boldsymbol{x},\boldsymbol{x}) \\
&\quad + \Bigl(\mathcal{U}^{\varepsilon,2}_{t,0}(\boldsymbol{x})\mathcal{P}^\varepsilon\mathcal{U}^{\varepsilon,0}_{0,t}(\boldsymbol{x})\Bigr)(\boldsymbol{x},\boldsymbol{x}).
\end{aligned}
$$

Therefore the density is given by

$$
\rho^\varepsilon(t,\boldsymbol{x}) = \rho^\varepsilon_0(t,\boldsymbol{x}) + \varepsilon\rho^\varepsilon_1(t,\boldsymbol{x}) + \varepsilon^2\rho^\varepsilon_2(t,\boldsymbol{x}) + \mathcal{O}(\varepsilon^3).
$$

The current is given by

$$\boldsymbol{J}^\varepsilon(t,\boldsymbol{x}) = \frac{1}{2i}\big[\varepsilon\nabla_{\boldsymbol{y}},\ \boldsymbol{\rho}^\varepsilon(t)\big]_+(\boldsymbol{x},\boldsymbol{x}),$$

where $\boldsymbol{\rho}^\varepsilon(t)$ is the density matrix in (4.17) and $[a,\ b]_+ = ab+ba$ is the anticommutator.

With the help of (4.23), similarly one has

$$\boldsymbol{J}^\varepsilon(t,\boldsymbol{x}) = \boldsymbol{J}_0^\varepsilon(t,\boldsymbol{x}) + \varepsilon\boldsymbol{J}_1^\varepsilon(t,\boldsymbol{x}) + \mathcal{O}(\varepsilon^2),$$

where

$$\boldsymbol{J}_0^\varepsilon = \frac{1}{2i}\left[\varepsilon\nabla_{\boldsymbol{y}},\ \mathcal{U}_{t,0}^{\varepsilon,0}(\boldsymbol{x})\mathcal{P}^\varepsilon\mathcal{U}_{0,t}^{\varepsilon,0}(\boldsymbol{x})\right]_+(\boldsymbol{x},\boldsymbol{x}),$$
$$\boldsymbol{J}_1^\varepsilon = \frac{1}{2i}\left[\varepsilon\nabla_{\boldsymbol{y}},\ \mathcal{U}_{t,0}^{\varepsilon,0}(\boldsymbol{x})\mathcal{P}^\varepsilon\mathcal{U}_{0,t}^{1,\varepsilon}(\boldsymbol{x}) + \mathcal{U}_{t,0}^{\varepsilon,1}(\boldsymbol{x})\mathcal{P}^\varepsilon\mathcal{U}_{0,t}^{\varepsilon,0}(\boldsymbol{x})\right]_+(\boldsymbol{x},\boldsymbol{x}) - \rho_0^\varepsilon\boldsymbol{A}_0(t,\boldsymbol{x}).$$

4.3. **Rescalings of Hamiltonian, density and current.** Notice that the operator $H_0^\varepsilon(t,\boldsymbol{x})$ in (4.19) can be rescaled as

$$\delta_\varepsilon H_0^\varepsilon(t,\boldsymbol{x})\delta_\varepsilon^* = H_0(t,\boldsymbol{x}),$$

with δ_ε as the dilation operator and H_0 is given by

$$H_0(t,\boldsymbol{x}) = -\tfrac{1}{2}\Delta_{\boldsymbol{\zeta}} + v_0(\boldsymbol{\zeta}) + U_0(t,\boldsymbol{x}),$$

where $\boldsymbol{\zeta} = \boldsymbol{y}/\varepsilon$ is the small scale spatial variable.

Therefore, we can rescale the expressions of $\rho_j^\varepsilon(t,\boldsymbol{x})$ and $\boldsymbol{J}_j^\varepsilon(t,\boldsymbol{x})$ by

$$\rho_j^\varepsilon(t,\boldsymbol{x}) = \varepsilon^{-3}\rho_j(t,\boldsymbol{x},\boldsymbol{x}/\varepsilon),\quad \boldsymbol{J}_j^\varepsilon(t,\boldsymbol{x}) = \varepsilon^{-3}\boldsymbol{J}_j(t,\boldsymbol{x},\boldsymbol{x}/\varepsilon),\quad j=0,1,\cdots,$$

and

$$\rho_0(t,\boldsymbol{x},\boldsymbol{z}) = \Big(\mathcal{U}_{t,0}^0(\boldsymbol{x})\mathcal{P}\mathcal{U}_{0,t}^0(\boldsymbol{x})\Big)(\boldsymbol{z},\boldsymbol{z}), \tag{4.24}$$

$$\rho_1(t,\boldsymbol{x},\boldsymbol{z}) = \Big(\mathcal{U}_{t,0}^0(\boldsymbol{x})\mathcal{P}\mathcal{U}_{0,t}^1(\boldsymbol{x},\boldsymbol{z})\Big)(\boldsymbol{z},\boldsymbol{z}) + \Big(\mathcal{U}_{t,0}^1(\boldsymbol{x},\boldsymbol{z})\mathcal{P}\mathcal{U}_{0,t}^0(\boldsymbol{x})\Big)(\boldsymbol{z},\boldsymbol{z}), \tag{4.25}$$

$$\begin{aligned}\rho_2(t,\boldsymbol{x},\boldsymbol{z}) = {}&\Big(\mathcal{U}_{t,0}^0(\boldsymbol{x})\mathcal{P}\mathcal{U}_{0,t}^2(\boldsymbol{x},\boldsymbol{z})\Big)(\boldsymbol{z},\boldsymbol{z}) + \Big(\mathcal{U}_{t,0}^1(\boldsymbol{x},\boldsymbol{z})\mathcal{P}\mathcal{U}_{0,t}^1(\boldsymbol{x},\boldsymbol{z})\Big)(\boldsymbol{z},\boldsymbol{z})\\ &+ \Big(\mathcal{U}_{t,0}^2(\boldsymbol{x},\boldsymbol{z})\mathcal{P}\mathcal{U}_{0,t}^0(\boldsymbol{x})\Big)(\boldsymbol{z},\boldsymbol{z}),\end{aligned} \tag{4.26}$$

$$\boldsymbol{J}_0(t,\boldsymbol{x},\boldsymbol{z}) = \frac{1}{2i}\left[\nabla_{\boldsymbol{\zeta}},\ \mathcal{U}_{t,0}^0(\boldsymbol{x})\mathcal{P}\mathcal{U}_{0,t}^0(\boldsymbol{x})\right](\boldsymbol{z},\boldsymbol{z}), \tag{4.27}$$

$$\begin{aligned}\boldsymbol{J}_1(t,\boldsymbol{x},\boldsymbol{z}) = {}&\frac{1}{2i}\left[\nabla_{\boldsymbol{\zeta}},\ \mathcal{U}_{t,0}^0(\boldsymbol{x})\mathcal{P}\mathcal{U}_{0,t}^1(\boldsymbol{x},\boldsymbol{z}) + \mathcal{U}_{t,0}^1(\boldsymbol{x},\boldsymbol{z})\mathcal{P}\mathcal{U}_{0,t}^0(\boldsymbol{x})\right](\boldsymbol{z},\boldsymbol{z})\\ &- \rho_0(t,\boldsymbol{x},\boldsymbol{z})\boldsymbol{A}_0(t,\boldsymbol{x}),\end{aligned} \tag{4.28}$$

where $\mathcal{P}$ is the rescaled (lattice parameter 1) density matrix for the unperturbed system, and $\mathcal{U}_{t,s}^i$ are propagation operators defined on $L^2_{\boldsymbol{\zeta}}$ by

$$\mathcal{U}_{t,s}^0(\boldsymbol{x}) = \mathcal{T}\exp(-i\int_s^t H_0(\tau,\boldsymbol{x})\,\mathrm{d}\tau),$$

$$\mathcal{U}^1_{t,s}(\boldsymbol{x},\boldsymbol{z}) = -i\int_s^t \mathcal{U}^0_{t,\tau}(\boldsymbol{x})\delta H_1(\tau,\boldsymbol{x},\boldsymbol{z})\mathcal{U}^0_{\tau,0}(\boldsymbol{x})\,\mathrm{d}\tau, \tag{4.29}$$

$$\begin{aligned}\mathcal{U}^2_{t,s}(\boldsymbol{x},\boldsymbol{z}) = &-i\int_s^t \mathcal{U}^0_{t,\tau}(\boldsymbol{x})\delta H_2(\tau,\boldsymbol{x},\boldsymbol{z})\mathcal{U}^0_{\tau,0}(\boldsymbol{x})\,\mathrm{d}\tau \\ &- \int_s^t\int_s^{\tau_2} \mathcal{U}^0_{t,\tau_2}(\boldsymbol{x})\delta H_1(\tau_2,\boldsymbol{x},\boldsymbol{z})\mathcal{U}^0_{\tau_2,\tau_1}(\boldsymbol{x})\delta H_1(\tau_1,\boldsymbol{x},\boldsymbol{z})\mathcal{U}^0_{\tau_1,0}(\boldsymbol{x})\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2,\end{aligned} \tag{4.30}$$

in which

$$H_0(t,\boldsymbol{x}) = -\tfrac{1}{2}\Delta_{\boldsymbol{\zeta}} + v_0(\boldsymbol{\zeta}) + U_0(t,\boldsymbol{x}), \tag{4.31}$$

$$\delta H_1(t,\boldsymbol{x},\boldsymbol{z}) = (\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}U_0(t,\boldsymbol{x}) + v_1(t,\boldsymbol{x},\boldsymbol{\zeta}) + U_1(t,\boldsymbol{x}) + i\boldsymbol{A}_0\cdot\nabla_{\boldsymbol{\zeta}}, \tag{4.32}$$

$$\begin{aligned}\delta H_2(t,\boldsymbol{x},\boldsymbol{z}) = &\tfrac{1}{2}\big((\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}\big)^2 U_0(t,\boldsymbol{x}) \\ &+ (\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}\big(v_1(t,\boldsymbol{x},\boldsymbol{\zeta}) + U_1(t,\boldsymbol{x})\big) + V_2(t,\boldsymbol{x},\boldsymbol{\zeta}) \\ &+ i\bigg((\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}\boldsymbol{A}_0 + \boldsymbol{A}_1\bigg)\cdot\nabla_{\boldsymbol{\zeta}} + \tfrac{1}{2}\left|\boldsymbol{A}_0\right|^2.\end{aligned} \tag{4.33}$$

Notice that the dependence of the operator $H_0(\tau,\boldsymbol{x})$ on $\boldsymbol{x}$ lies only in $U(\tau,\boldsymbol{x})$ which works like a number as an operator on $L^2_{\boldsymbol{\zeta}}$. It implies

$$\mathcal{U}^0_{t,s}(\boldsymbol{x}) = \exp\bigg(-i\int_s^t U(\tau,\boldsymbol{x})\bigg)\exp(-i(t-s)H_0),$$

where H_0 agrees with the unperturbed Hamiltonian

$$H_0 = -\tfrac{1}{2}\Delta_{\boldsymbol{\zeta}} + v_0(\boldsymbol{\zeta}). \tag{4.34}$$

Moreover, in the expression of ρ_j, the phase factor $\exp(-i\int_s^t U(\tau,\boldsymbol{x}))$ will not appear since it gets canceled by its complex conjugate. Therefore, we can simply take

$$\mathcal{U}^0_{t,s} = \exp(-i(t-s)H_0), \tag{4.35}$$

which is, in particular, independent of $\boldsymbol{x}$.

It results that the leading order density agrees with the ground state electron density of the unperturbed system,

$$\rho_0(t,\boldsymbol{x},\boldsymbol{z}) = e^{-itH_0}\mathcal{P}e^{itH_0}(\boldsymbol{z},\boldsymbol{z}) = \rho_{\mathrm{gs}}(\boldsymbol{z}). \tag{4.36}$$

This also shows the following proposition.

Proposition 4.1. *ρ_0 is independent of t and $\boldsymbol{x}$ and*

$$\langle\rho_0\rangle_{\boldsymbol{z}} = \langle m_0\rangle_{\boldsymbol{z}},$$

which satisfies the first constraint of (4.13) *self-consistently.*

5. Effective equations in the time domain

In this section, we derive the main result in time domain. It will be connected to the main result described in frequency domain in the next Section.

5.1. **Effective equations in time domain.** Let us first summarize the resulting equations in the time domain. Recall that

$$\begin{aligned} V_0(t,\boldsymbol{x},\boldsymbol{z}) &= v_0(\boldsymbol{z}) + U_0(t,\boldsymbol{x}),\\ V_1(t,\boldsymbol{x},\boldsymbol{z}) &= v_1(t,\boldsymbol{x},\boldsymbol{z}) + U_1(t,\boldsymbol{x}),\\ \boldsymbol{A}_0(t,\boldsymbol{x},\boldsymbol{z}) &= \boldsymbol{A}_0(t,\boldsymbol{x}), \end{aligned}$$

then one has the following effective equations from (3.18)-(3.22).

- The microscopic scalar potential $v_0(\boldsymbol{z})$ is the same as that of the unperturbed system.
- The potentials $v_1(t,\boldsymbol{x},\boldsymbol{z})$, $U_0(t,\boldsymbol{x})$ and $\boldsymbol{A}_0(t,\boldsymbol{x})$ form a closed system as described below. The microscopic scalar potential $v_1(t,\boldsymbol{x},\boldsymbol{z})$ is given by

$$\begin{aligned} v_1 &= V_1(t,\boldsymbol{x},\boldsymbol{z}) - \langle V_1(t,\boldsymbol{x},\boldsymbol{z})\rangle_{\boldsymbol{z}},\\ V_1 &= \mathcal{V}\rho_1 = \phi_1 + \eta'(\rho_0)\rho_1, \\ -\Delta_{\boldsymbol{z}}\phi_1 &= \rho_1. \end{aligned} \tag{5.1}$$

Recall that $\mathcal{V}$ is the linearization of the effective potential operator at equilibrium density. Here, the density ρ_1 is given by the equation

$$(I-\chi\mathcal{V})\rho_1 = \int_0^t \boldsymbol{f}(t-\tau)\cdot\nabla_{\boldsymbol{x}}U_0(\tau)\,\mathrm{d}\tau + \int_0^t \boldsymbol{g}(t-\tau)\cdot\boldsymbol{A}_0(\tau)\,\mathrm{d}\tau, \tag{5.2}$$

where I is the identity operator,

$$\begin{aligned} \chi\mathcal{V}\rho_1 = \chi v_1 = -2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{\Gamma^*} & e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)} \\ &\times u_{n,\boldsymbol{k}}u_{m,\boldsymbol{k}}^*\langle u_{n,\boldsymbol{k}}|v_1(\tau)|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\tau, \end{aligned}$$

and

$$\begin{aligned} \boldsymbol{f}(s) &= -2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\fint_{\Gamma^*} e^{i\omega_{mn}(\boldsymbol{k})s}u_{n,\boldsymbol{k}}u_{m,\boldsymbol{k}}^*\langle u_{n,\boldsymbol{k}}|i\nabla_{\boldsymbol{k}}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\,\mathrm{d}\boldsymbol{k},\\ \boldsymbol{g}(s) &= -2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\fint_{\Gamma^*} e^{i\omega_{mn}(\boldsymbol{k})s}u_{n,\boldsymbol{k}}u_{m,\boldsymbol{k}}^*\langle u_{n,\boldsymbol{k}}|i\nabla_{\boldsymbol{\zeta}}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\,\mathrm{d}\boldsymbol{k}. \end{aligned}$$

Here we have introduced the short hand notation

$$\omega_{mn}(\boldsymbol{k}) = E_m(\boldsymbol{k}) - E_n(\boldsymbol{k}).$$

- The macroscopic scalar potential $U_0(t,\boldsymbol{x})$ satisfies

$$-\Delta U_0 - P_{\alpha\beta}\partial_{\boldsymbol{x}_\alpha}\partial_{\boldsymbol{x}_\beta}U_0 = Q_\alpha\left(\partial_{\boldsymbol{x}_\alpha}v_1\right) + R_{\alpha\beta}\big(\partial_{\boldsymbol{x}_\alpha}(\boldsymbol{A}_0)_\beta\big) + \rho_{\mathrm{ext}}(t,\boldsymbol{x}). \tag{5.3}$$

Here

$$\begin{aligned} P_{\alpha\beta}\left(\partial_{\boldsymbol{x}_\alpha}\partial_{\boldsymbol{x}_\beta}U_0\right) = 2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\int_0^t\fint_{\Gamma^*} & e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)}\overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)} \\ &\times\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\partial_{\boldsymbol{x}_\alpha}\partial_{\boldsymbol{x}_\beta}U_0(\tau)\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\tau, \end{aligned}$$

$$Q_\alpha\left(\partial_{\boldsymbol{x}_\alpha} v_1\right) = 2\mathfrak{Im} \sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{\Gamma^*} e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)}\overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)} \times \langle u_{n,\boldsymbol{k}}|\partial_{\boldsymbol{x}_\alpha} v_1(\tau)|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\tau,$$

and

$$R_{\alpha\beta}\big(\partial_{\boldsymbol{x}_\alpha}(\boldsymbol{A}_0)_\beta\big) = 2\mathfrak{Im} \sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{\Gamma^*} e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)}\overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)} \times \langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{\zeta}_\beta}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\partial_{\boldsymbol{x}_\alpha}(\boldsymbol{A}_0)_\beta\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\tau.$$

- The macroscopic vector potential $\boldsymbol{A}_0$ satisfies

$$\begin{aligned}&\frac{\partial^2}{\partial t^2}(\boldsymbol{A}_0)_\alpha - \Delta_{\boldsymbol{x}}(\boldsymbol{A}_0)_\alpha + \frac{\partial}{\partial t}(\partial_{\boldsymbol{x}_\alpha}U_0) = \\ &\qquad S_\alpha(v_1) + M_{\alpha\beta}(\partial_{\boldsymbol{x}_\beta}U_0) + N_{\alpha\beta}\big((\boldsymbol{A}_0)_\beta\big) - \langle\rho_0\rangle_z(\boldsymbol{A}_0)_\alpha + (\boldsymbol{J}_{\text{ext}})_\alpha(t,\boldsymbol{x}),\\ &\nabla_{\boldsymbol{x}}\cdot\boldsymbol{A}_0 = 0.\end{aligned}\tag{5.4}$$

Here

$$S_\alpha(v_1) = 2\mathfrak{Im} \sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{\Gamma^*} e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)}\overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{\zeta}_\alpha}|u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)} \times \langle u_{n,\boldsymbol{k}}|v_1(\tau)|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\tau,$$

$$M_{\alpha\beta}\left(\partial_{\boldsymbol{x}_\beta}U_0\right) = 2\mathfrak{Im} \sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{\Gamma^*} e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)}\overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{\zeta}_\alpha}|u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)} \times \langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\partial_{\boldsymbol{x}_\beta}U_0(\tau)\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\tau,$$

and

$$N_{\alpha\beta}\big((\boldsymbol{A}_0)_\beta\big) = 2\mathfrak{Im} \sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{\Gamma^*} e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)}\overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{\zeta}_\alpha}|u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)} \times \langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{\zeta}_\beta}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}(\boldsymbol{A}_0)_\beta(\tau)\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\tau.$$

The above equations from (5.1) to (5.4) form a *closed* system that determines the macroscopic potentials $U_0(t,\boldsymbol{x})$ and $\boldsymbol{A}_0(t,\boldsymbol{x})$.

5.2. **Derivation.** We first give a description on how to derive the equations (5.1)-(5.4). By taking the $\boldsymbol{z}$-average of (4.7) and (4.12), one can get the effective equations for $U_0(t,\boldsymbol{x})$ and $\boldsymbol{A}_0(t,\boldsymbol{x})$. However, in order to get equations in explicit form, one needs the expressions of ρ_1, $\langle\rho_2\rangle_{\boldsymbol{z}}$, $\boldsymbol{J}_0$ and $\langle\boldsymbol{J}_1\rangle_{\boldsymbol{z}}$ in terms of Bloch wave function $\{\psi_{n,\boldsymbol{k}}\}$ or its periodic part $\{u_{n,\boldsymbol{k}}\}$. This will require the following three lemmas. The first lemma states the property of perturbed density under Hamiltonian perturbation, and the other two introduce some identities related with Bloch waves. These identities will be useful in simplifying the expressions of density and current.

Lemma 5.1. *We consider the electron dynamics under the perturbed Hamiltonian* $\widetilde{H} = H_0 + \sum_{j=1}^{J} \varepsilon^j (V_j + i\boldsymbol{A}_{j-1} \cdot \nabla_{\boldsymbol{\zeta}})$ *and denote the density as*

$$\widetilde{\rho}(t, \boldsymbol{x}, \boldsymbol{z}) = \rho_0(t, \boldsymbol{x}, \boldsymbol{z}) + \sum_k \varepsilon^k \widetilde{\rho}_k(t, \boldsymbol{x}, \boldsymbol{z}).$$

Assume the initial condition is

$$\widetilde{\rho}(0, \boldsymbol{x}, \boldsymbol{z}) = \rho_{\mathrm{gs}}(\boldsymbol{z}),$$

then one has for any k*,*

$$\langle \widetilde{\rho}_k(t, \boldsymbol{x}, \boldsymbol{z}) \rangle_{\boldsymbol{z}} = 0.$$

Proof. Since we choose the Coulomb gauge so that $\nabla_{\boldsymbol{\zeta}} \cdot \boldsymbol{A}_{j-1} = 0,\ j = 1, \cdots, J$, the operator $\mathcal{T} \exp\big(-i \int_0^t \widetilde{H}(\tau)\, \mathrm{d}\tau\big)$ is unitary, which produces

$$\langle \widetilde{\rho}(t, \boldsymbol{x}, \boldsymbol{z}) \rangle_{\boldsymbol{z}} = \langle \widetilde{\rho}(0, \boldsymbol{x}, \boldsymbol{z}) \rangle_{\boldsymbol{z}} = \langle \rho_{\mathrm{gs}}(\boldsymbol{z}) \rangle_{\boldsymbol{z}}.$$

Therefore by (4.36) and

$$\langle \widetilde{\rho}(t, \boldsymbol{x}, \boldsymbol{z}) \rangle_{\boldsymbol{z}} = \langle \rho_0(t, \boldsymbol{x}, \boldsymbol{z}) \rangle_{\boldsymbol{z}} + \sum_k \varepsilon^k \langle \widetilde{\rho}_k(t, \boldsymbol{x}, \boldsymbol{z}) \rangle_{\boldsymbol{z}},$$

one gets, for any k,

$$\langle \widetilde{\rho}_k(t, \boldsymbol{x}, \boldsymbol{z}) \rangle_{\boldsymbol{z}} = 0.$$

□

Lemma 5.2. *For any* $n, m \in \mathbb{Z}_+$ *and* $\boldsymbol{k}, \boldsymbol{p} \in \Gamma^*$*, the following equations hold in the distributional sense.*

$$\int_{\mathbb{R}^3} (\boldsymbol{\zeta} - \boldsymbol{z}) \psi^*_{n,\boldsymbol{k}} \psi_{m,\boldsymbol{p}}\, \mathrm{d}\boldsymbol{\zeta} = \Big\{ \delta_{nm}(-\boldsymbol{z} - i\partial_{\boldsymbol{p}}) + \langle u_{n,\boldsymbol{k}} | i\nabla_{\boldsymbol{k}} | u_{m,\boldsymbol{k}} \rangle_{L^2(\Gamma)} \Big\} \delta(\boldsymbol{p} - \boldsymbol{k})\, |\Gamma^*|\,, \tag{5.5}$$

$$\int_{\mathbb{R}^3} v_1(\tau, \boldsymbol{x}, \boldsymbol{\zeta}) \psi^*_{n,\boldsymbol{k}} \psi_{m,\boldsymbol{p}}\, \mathrm{d}\boldsymbol{\zeta} = \langle u_{n,\boldsymbol{k}} | v_1(\tau) | u_{m,\boldsymbol{k}} \rangle_{L^2(\Gamma)} \delta(\boldsymbol{p} - \boldsymbol{k})\, |\Gamma^*|\,, \tag{5.6}$$

$$\int_{\mathbb{R}^3} \psi^*_{n,\boldsymbol{k}} (i\nabla_{\boldsymbol{\zeta}}) \psi_{m,\boldsymbol{p}}\, \mathrm{d}\boldsymbol{\zeta} = \langle u_{n,\boldsymbol{k}} | i\nabla_{\boldsymbol{\zeta}} | u_{m,\boldsymbol{k}} \rangle_{L^2(\Gamma)} \delta(\boldsymbol{p} - \boldsymbol{k})\, |\Gamma^*|\,. \tag{5.7}$$

Proof. By the definition of Bloch wave, direct calculation yields

$$\begin{aligned}
\int_{\mathbb{R}^3} (\boldsymbol{\zeta} - \boldsymbol{z}) \psi^*_{n,\boldsymbol{k}} \psi_{m,\boldsymbol{p}}\, \mathrm{d}\boldsymbol{\zeta} &= \int_{\mathbb{R}^3} (\boldsymbol{\zeta} - \boldsymbol{z}) e^{i\boldsymbol{\zeta} \cdot (\boldsymbol{p} - \boldsymbol{k})} u^*_{n,\boldsymbol{k}} u_{m,\boldsymbol{p}}\, \mathrm{d}\boldsymbol{\zeta} \\
&= \sum_{\boldsymbol{X}_j \in \mathbb{L}} \int_\Gamma (\boldsymbol{\zeta} + \boldsymbol{X}_j - \boldsymbol{z}) e^{i(\boldsymbol{\zeta} + \boldsymbol{X}_j) \cdot (\boldsymbol{p} - \boldsymbol{k})} u^*_{n,\boldsymbol{k}} u_{m,\boldsymbol{p}}\, \mathrm{d}\boldsymbol{\zeta} \\
&= \sum_{\boldsymbol{X}_j \in \mathbb{L}} \int_\Gamma \Big((-i\partial_{\boldsymbol{p}} - \boldsymbol{z}) e^{i(\boldsymbol{\zeta} + \boldsymbol{X}_j) \cdot (\boldsymbol{p} - \boldsymbol{k})} \Big) u^*_{n,\boldsymbol{k}} u_{m,\boldsymbol{p}}\, \mathrm{d}\boldsymbol{\zeta}.
\end{aligned}$$

For the right hand side, we have

$$
\begin{aligned}
\sum_{\boldsymbol{X}_j\in\mathbb{L}} \int_\Gamma \Big(-i\partial_{\boldsymbol{p}} e^{i(\boldsymbol{\zeta}+\boldsymbol{X}_j)\cdot(\boldsymbol{p}-\boldsymbol{k})}\Big) u_{n,\boldsymbol{k}}^* u_{m,\boldsymbol{p}} \,\mathrm{d}\boldsymbol{\zeta} = & \\
& - i\partial_{\boldsymbol{p}} \Big(\sum_{\boldsymbol{X}_j\in\mathbb{L}} \int_\Gamma e^{i(\boldsymbol{\zeta}+\boldsymbol{X}_j)\cdot(\boldsymbol{p}-\boldsymbol{k})} u_{n,\boldsymbol{k}}^* u_{m,\boldsymbol{p}} \,\mathrm{d}\boldsymbol{\zeta} \Big) \\
& + \sum_{\boldsymbol{X}_j\in\mathbb{L}} \int_\Gamma e^{i(\boldsymbol{\zeta}+\boldsymbol{X}_j)\cdot(\boldsymbol{p}-\boldsymbol{k})} u_{n,\boldsymbol{k}}^* (i\partial_{\boldsymbol{p}}) u_{m,\boldsymbol{p}} \,\mathrm{d}\boldsymbol{\zeta} \\
= & - i\partial_{\boldsymbol{p}} \Big(\sum_{\boldsymbol{X}_j\in\mathbb{L}} e^{i\boldsymbol{X}_j\cdot(\boldsymbol{p}-\boldsymbol{k})} \int_\Gamma e^{i\boldsymbol{\zeta}\cdot(\boldsymbol{p}-\boldsymbol{k})} u_{n,\boldsymbol{k}}^* u_{m,\boldsymbol{p}} \,\mathrm{d}\boldsymbol{\zeta} \Big) \\
& + \sum_{\boldsymbol{X}_j\in\mathbb{L}} e^{i\boldsymbol{X}_j\cdot(\boldsymbol{p}-\boldsymbol{k})} \int_\Gamma e^{i\boldsymbol{\zeta}\cdot(\boldsymbol{p}-\boldsymbol{k})} u_{n,\boldsymbol{k}}^* (i\partial_{\boldsymbol{p}}) u_{m,\boldsymbol{p}} \,\mathrm{d}\boldsymbol{\zeta}.
\end{aligned} \tag{5.8}
$$

To further simplify the above expression, we use the Poisson summation formula

$$
\sum_{\boldsymbol{X}_j\in\mathbb{L}} e^{i\boldsymbol{X}_j\cdot\boldsymbol{k}} = |\Gamma^*| \sum_{\boldsymbol{K}_j\in\mathbb{L}^*} \delta(\boldsymbol{k}-\boldsymbol{K}_j), \tag{5.9}
$$

in the distributional sense. Substitute (5.9) into (5.8), we obtain

$$
\begin{aligned}
\sum_{\boldsymbol{X}_j\in\mathbb{L}} \int_\Gamma \Big(-i\partial_{\boldsymbol{p}} e^{i(\boldsymbol{\zeta}+\boldsymbol{X}_j)\cdot(\boldsymbol{p}-\boldsymbol{k})}\Big) u_{n,\boldsymbol{k}}^* u_{m,\boldsymbol{p}} \,\mathrm{d}\boldsymbol{\zeta} = & \\
& - |\Gamma^*|\, i\partial_{\boldsymbol{p}} \Big(\delta(\boldsymbol{p}-\boldsymbol{k}) \int_\Gamma e^{i\boldsymbol{\zeta}\cdot(\boldsymbol{p}-\boldsymbol{k})} u_{n,\boldsymbol{k}}^* u_{m,\boldsymbol{p}} \,\mathrm{d}\boldsymbol{\zeta} \Big) \\
& + |\Gamma^*|\, \delta(\boldsymbol{p}-\boldsymbol{k}) \int_\Gamma e^{i\boldsymbol{\zeta}\cdot(\boldsymbol{p}-\boldsymbol{k})} u_{n,\boldsymbol{k}}^* (i\partial_{\boldsymbol{p}}) u_{m,\boldsymbol{p}} \,\mathrm{d}\boldsymbol{\zeta} \\
= & - |\Gamma^*|\, i\partial_{\boldsymbol{p}} \Big(\delta(\boldsymbol{p}-\boldsymbol{k}) \int_\Gamma e^{i\boldsymbol{\zeta}\cdot(\boldsymbol{p}-\boldsymbol{k})} u_{n,\boldsymbol{k}}^* u_{m,\boldsymbol{p}} \,\mathrm{d}\boldsymbol{\zeta} \Big) \\
& + |\Gamma^*|\, \delta(\boldsymbol{p}-\boldsymbol{k}) \langle u_{n,\boldsymbol{k}} | i\nabla_{\boldsymbol{k}} | u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \\
= & - |\Gamma^*|\, i\partial_{\boldsymbol{p}} \Big(\delta(\boldsymbol{p}-\boldsymbol{k}) \langle u_{n,\boldsymbol{k}} | u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \Big) \\
& + |\Gamma^*|\, \delta(\boldsymbol{p}-\boldsymbol{k}) \langle u_{n,\boldsymbol{k}} | i\nabla_{\boldsymbol{k}} | u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \\
= & |\Gamma^*|\, (-i\partial_{\boldsymbol{p}}) \delta(\boldsymbol{p}-\boldsymbol{k}) \delta_{mn} + |\Gamma^*|\, \delta(\boldsymbol{p}-\boldsymbol{k}) \langle u_{n,\boldsymbol{k}} | i\nabla_{\boldsymbol{k}} | u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}.
\end{aligned}
$$

The last equality follows from the orthogonality of $\{u_{n,\boldsymbol{k}}\}$ for each $\boldsymbol{k}$.

Similarly we have

$$
\begin{aligned}
\sum_{\boldsymbol{X}_j\in\mathbb{L}} \int_\Gamma -\boldsymbol{z} e^{i(\boldsymbol{\zeta}+\boldsymbol{X}_j)\cdot(\boldsymbol{p}-\boldsymbol{k})} u_{n,\boldsymbol{k}}^* u_{m,\boldsymbol{p}} \,\mathrm{d}\boldsymbol{\zeta} & \\
& = -\boldsymbol{z}\, |\Gamma^*|\, \delta(\boldsymbol{p}-\boldsymbol{k}) \int_\Gamma e^{i\boldsymbol{\zeta}\cdot(\boldsymbol{p}-\boldsymbol{k})} u_{n,\boldsymbol{k}}^* u_{m,\boldsymbol{p}} \,\mathrm{d}\boldsymbol{\zeta} \\
& = -\boldsymbol{z}\, |\Gamma^*|\, \delta(\boldsymbol{p}-\boldsymbol{k}) \langle u_{n,\boldsymbol{k}} | u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \\
& = -\boldsymbol{z}\, |\Gamma^*|\, \delta(\boldsymbol{p}-\boldsymbol{k}) \delta_{mn},
\end{aligned} \tag{5.10}
$$

and

$$\begin{aligned}\int_{\mathbb{R}^3} v_1(\tau,\boldsymbol{x},\boldsymbol{\zeta})\psi^*_{n,\boldsymbol{k}}\psi_{m,\boldsymbol{p}}\,\mathrm{d}\boldsymbol{\zeta} &= \int_{\mathbb{R}^3} v_1(\tau,\boldsymbol{x},\boldsymbol{\zeta})e^{i\boldsymbol{\zeta}\cdot(\boldsymbol{p}-\boldsymbol{k})}u^*_{n,\boldsymbol{k}}u_{m,\boldsymbol{p}}\,\mathrm{d}\boldsymbol{\zeta}\\ &= \sum_{\boldsymbol{X}_j\in\mathbb{L}}\int_\Gamma v_1(\tau,\boldsymbol{x},\boldsymbol{\zeta})e^{i(\boldsymbol{\zeta}+\boldsymbol{X}_j)\cdot(\boldsymbol{p}-\boldsymbol{k})}u^*_{n,\boldsymbol{k}}u_{m,\boldsymbol{p}}\,\mathrm{d}\boldsymbol{\zeta}\\ &= \langle u_{n,\boldsymbol{k}}|v_1(\tau)|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\delta(\boldsymbol{p}-\boldsymbol{k})\,|\Gamma^*|\,,\end{aligned}$$

where we have used the periodicity of $v_1(\tau,\boldsymbol{x},\boldsymbol{\zeta})$ in $\boldsymbol{\zeta}$.

Hence combining the above equations together yields (5.5), and the last equality proves (5.6). The proof of (5.7) is essentially the same as (5.6) which we will omit here. □

Lemma 5.3. *For any $n,m\in\mathbb{Z}_+$ and $\boldsymbol{k},\boldsymbol{p}\in\Gamma^*$, we have in the distributional sense,*

$$\begin{aligned}&\int_{\mathbb{R}^3}(\boldsymbol{\zeta}-\boldsymbol{z})_\alpha(\boldsymbol{\zeta}-\boldsymbol{z})_\beta\psi^*_{n,\boldsymbol{k}}\psi_{m,\boldsymbol{p}}\,\mathrm{d}\boldsymbol{\zeta} =\\ &\qquad \delta_{nm}(\boldsymbol{z}_\alpha\boldsymbol{z}_\beta-\partial_{\boldsymbol{p}_\alpha}\partial_{\boldsymbol{p}_\beta})\delta(\boldsymbol{p}-\boldsymbol{k})\,|\Gamma^*|\\ &\qquad + \langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}(-i\boldsymbol{z}_\alpha-i\partial_{\boldsymbol{p}_\alpha})\delta(\boldsymbol{p}-\boldsymbol{k})\,|\Gamma^*|\\ &\qquad + \langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}(-i\boldsymbol{z}_\beta-i\partial_{\boldsymbol{p}_\beta})\delta(\boldsymbol{p}-\boldsymbol{k})\,|\Gamma^*|\\ &\qquad - \langle u_{n,\boldsymbol{k}}|\partial_{\boldsymbol{k}_\alpha}\partial_{\boldsymbol{k}_\beta}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\delta(\boldsymbol{p}-\boldsymbol{k})\,|\Gamma^*|\,.\end{aligned}\tag{5.11}$$

$$\begin{aligned}&\int_{\mathbb{R}^3}(\boldsymbol{\zeta}-\boldsymbol{z})_\alpha\partial_{\boldsymbol{x}_\alpha}v_1(\tau,\boldsymbol{x},\boldsymbol{\zeta})\psi^*_{n,\boldsymbol{k}}\psi_{m,\boldsymbol{p}}\,\mathrm{d}\boldsymbol{\zeta} =\\ &\qquad \langle u_{n,\boldsymbol{k}}|\partial_{\boldsymbol{x}_\alpha}v_1 i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\delta(\boldsymbol{p}-\boldsymbol{k})\,|\Gamma^*|\\ &\qquad + \langle u_{n,\boldsymbol{k}}|\partial_{\boldsymbol{x}_\alpha}v_1|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}(-\boldsymbol{z}_\alpha-i\partial_{\boldsymbol{p}_\alpha})\delta(\boldsymbol{p}-\boldsymbol{k})\,|\Gamma^*|\,.\end{aligned}\tag{5.12}$$

$$\begin{aligned}&\int_{\mathbb{R}^3}(\boldsymbol{\zeta}-\boldsymbol{z})_\alpha\psi^*_{n,\boldsymbol{k}}(i\partial_{\boldsymbol{\zeta}_\beta})\psi_{m,\boldsymbol{p}}\,\mathrm{d}\boldsymbol{\zeta} =\\ &\qquad \langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}(-\boldsymbol{k}_\beta+i\partial_{\boldsymbol{\zeta}_\beta})|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\delta(\boldsymbol{p}-\boldsymbol{k})\,|\Gamma^*|\\ &\qquad + \langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{\zeta}_\beta}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}(-\boldsymbol{z}_\alpha-i\partial_{\boldsymbol{p}_\alpha})\delta(\boldsymbol{p}-\boldsymbol{k})\,|\Gamma^*|\,.\end{aligned}\tag{5.13}$$

Proof. We calculate

$$\begin{aligned}\int_{\mathbb{R}^3}\boldsymbol{\zeta}_\alpha\boldsymbol{\zeta}_\beta\psi^*_{n,\boldsymbol{k}}\psi_{m,\boldsymbol{p}}\,\mathrm{d}\boldsymbol{\zeta} &= \int_{\mathbb{R}^3}\boldsymbol{\zeta}_\alpha\boldsymbol{\zeta}_\beta u^*_{n,\boldsymbol{k}}u_{m,\boldsymbol{p}}e^{i(\boldsymbol{p}-\boldsymbol{k})\boldsymbol{\zeta}}\,\mathrm{d}\boldsymbol{\zeta}\\ &= -\sum_{\boldsymbol{X}_j\in\mathbb{L}}\int_\Gamma u^*_{n,\boldsymbol{k}}u_{m,\boldsymbol{p}}\partial_{\boldsymbol{p}_\alpha}\partial_{\boldsymbol{p}_\beta}e^{i(\boldsymbol{p}-\boldsymbol{k})\cdot(\boldsymbol{\zeta}+\boldsymbol{X}_j)}\,\mathrm{d}\boldsymbol{\zeta}.\end{aligned}$$

The Leibniz rule gives

$$\begin{aligned}\int_{\mathbb{R}^3} \boldsymbol{\zeta}_\alpha \boldsymbol{\zeta}_\beta \psi^*_{n,\boldsymbol{k}} \psi_{m,\boldsymbol{p}} \,\mathrm{d}\boldsymbol{\zeta} = \\ &- \partial_{\boldsymbol{p}_\alpha} \partial_{\boldsymbol{p}_\beta} \Big(\int_\Gamma u^*_{n,\boldsymbol{k}} u_{m,\boldsymbol{p}} e^{i(\boldsymbol{p}-\boldsymbol{k})\cdot \boldsymbol{\zeta}} \,\mathrm{d}\boldsymbol{\zeta} \sum_{\boldsymbol{X}_j \in \mathbb{L}} e^{i(\boldsymbol{p}-\boldsymbol{k})\cdot \boldsymbol{X}_j} \Big) \\ &+ \partial_{\boldsymbol{p}_\alpha} \Big(\int_\Gamma u^*_{n,\boldsymbol{k}} (\partial_{\boldsymbol{p}_\beta} u_{m,\boldsymbol{p}}) e^{i(\boldsymbol{p}-\boldsymbol{k})\cdot \boldsymbol{\zeta}} \,\mathrm{d}\boldsymbol{\zeta} \sum_{\boldsymbol{X}_j \in \mathbb{L}} e^{i(\boldsymbol{p}-\boldsymbol{k})\cdot \boldsymbol{X}_j} \Big) \\ &+ \partial_{\boldsymbol{p}_\beta} \Big(\int_\Gamma u^*_{n,\boldsymbol{k}} (\partial_{\boldsymbol{p}_\alpha} u_{m,\boldsymbol{p}}) e^{i(\boldsymbol{p}-\boldsymbol{k})\cdot \boldsymbol{\zeta}} \,\mathrm{d}\boldsymbol{\zeta} \sum_{\boldsymbol{X}_j \in \mathbb{L}} e^{i(\boldsymbol{p}-\boldsymbol{k})\cdot \boldsymbol{X}_j} \Big) \\ &- \int_\Gamma u^*_{n,\boldsymbol{k}} (\partial_{\boldsymbol{p}_\alpha} \partial_{\boldsymbol{p}_\beta} u_{m,\boldsymbol{p}}) e^{i(\boldsymbol{p}-\boldsymbol{k})\cdot \boldsymbol{\zeta}} \,\mathrm{d}\boldsymbol{\zeta} \sum_{\boldsymbol{X}_j \in \mathbb{L}} e^{i(\boldsymbol{p}-\boldsymbol{k})\cdot \boldsymbol{X}_j}.\end{aligned}$$

Therefore applying the Poisson summation formula (5.9) yields

$$\begin{aligned}\int_{\mathbb{R}^3} \boldsymbol{\zeta}_\alpha \boldsymbol{\zeta}_\beta \psi^*_{n,\boldsymbol{k}} \psi_{m,\boldsymbol{p}} \,\mathrm{d}\boldsymbol{\zeta} =& - \delta_{nm} \partial_{\boldsymbol{p}_\alpha} \partial_{\boldsymbol{p}_\beta} \delta(\boldsymbol{p}-\boldsymbol{k}) \, |\Gamma^*| \\ &+ \langle u_{n,\boldsymbol{k}}, \partial_{\boldsymbol{k}_\beta} u_{m,\boldsymbol{k}} \rangle_{L^2(\Gamma)} \partial_{\boldsymbol{p}_\alpha} \delta(\boldsymbol{p}-\boldsymbol{k}) \, |\Gamma^*| \\ &+ \langle u_{n,\boldsymbol{k}}, \partial_{\boldsymbol{k}_\alpha} u_{m,\boldsymbol{k}} \rangle_{L^2(\Gamma)} \partial_{\boldsymbol{p}_\beta} \delta(\boldsymbol{p}-\boldsymbol{k}) \, |\Gamma^*| \\ &- \langle u_{n,\boldsymbol{k}}, \partial_{\boldsymbol{k}_\alpha} \partial_{\boldsymbol{k}_\beta} u_{m,\boldsymbol{k}} \rangle_{L^2(\Gamma)} \delta(\boldsymbol{p}-\boldsymbol{k}) \, |\Gamma^*| \,,\end{aligned}$$

and hence using (5.5) and (5.10), we have (5.11).

The calculations for (5.12) and (5.13) are analogous and omitted here.

□

5.2.1. *Derivation of the equation* (5.2). By (4.25), (4.35) and (4.29) the first order density perturbation reads as

$$\begin{aligned}\rho_1(t,\boldsymbol{x},\boldsymbol{z}) &= \Big(\mathcal{U}^0_{t,0}(\boldsymbol{x}) \mathcal{P} \mathcal{U}^1_{0,t}(\boldsymbol{x},\boldsymbol{z}) \Big)(\boldsymbol{z},\boldsymbol{z}) + \Big(\mathcal{U}^1_{t,0}(\boldsymbol{x},\boldsymbol{z}) \mathcal{P} \mathcal{U}^0_{0,t}(\boldsymbol{x}) \Big)(\boldsymbol{z},\boldsymbol{z}) \\ &= i e^{-itH_0} \mathcal{P} \int_0^t e^{i\tau H_0} \delta H_1(\tau,\boldsymbol{x},\boldsymbol{z}) e^{i(t-\tau)H_0} \,\mathrm{d}\tau(\boldsymbol{z},\boldsymbol{z}) + \text{c.c.},\end{aligned} \tag{5.14}$$

where we have used the fact that

$$\overline{(\mathcal{U}^1_{t,0}(\boldsymbol{x},\boldsymbol{z}) \mathcal{P} \mathcal{U}^0_{0,t}(\boldsymbol{x}))(\boldsymbol{z},\boldsymbol{z})} = (\mathcal{U}^0_{t,0}(\boldsymbol{x}) \mathcal{P} \mathcal{U}^1_{0,t}(\boldsymbol{x},\boldsymbol{z}))(\boldsymbol{z},\boldsymbol{z}),$$

as a direct consequence of

$$(\mathcal{U}^1_{t,0}(\boldsymbol{x},\boldsymbol{z}) \mathcal{P} \mathcal{U}^0_{0,t}(\boldsymbol{x}))^* = \mathcal{U}^0_{t,0}(\boldsymbol{x}) \mathcal{P} \mathcal{U}^1_{0,t}(\boldsymbol{x},\boldsymbol{z})$$

in the operator sense.

Substitute (4.32) into (5.14), we obtain

(5.15)
$$\begin{aligned}\rho_1(t,\boldsymbol{x},\boldsymbol{z}) =& -2\mathfrak{Im}\, e^{-itH_0}\mathcal{P}\int_0^t e^{i\tau H_0}(\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}U_0(\tau,\boldsymbol{x})e^{i(t-\tau)H_0}\,\mathrm{d}\tau(\boldsymbol{z},\boldsymbol{z})\\ &-2\mathfrak{Im}\, e^{-itH_0}\mathcal{P}\int_0^t e^{i\tau H_0}v_1(\tau,\boldsymbol{x},\boldsymbol{\zeta})e^{i(t-\tau)H_0}\,\mathrm{d}\tau(\boldsymbol{z},\boldsymbol{z}),\\ &-2\mathfrak{Im}\, e^{-itH_0}\mathcal{P}\int_0^t e^{i\tau H_0}\boldsymbol{A}_0(\tau,\boldsymbol{x})\cdot(i\nabla_{\boldsymbol{\zeta}})e^{i(t-\tau)H_0}\,\mathrm{d}\tau(\boldsymbol{z},\boldsymbol{z}),\end{aligned}$$

in getting which, we have used

$$\begin{aligned}&-2\mathfrak{Im}\, e^{-itH_0}\mathcal{P}\int_0^t e^{i\tau H_0}U_1(\tau,\boldsymbol{x})e^{i(t-\tau)H_0}\,\mathrm{d}\tau(\boldsymbol{z},\boldsymbol{z})\\ =&-2\mathfrak{Im}\, e^{-itH_0}\mathcal{P}e^{itH_0}(\boldsymbol{z},\boldsymbol{z})\int_0^t U_1(\tau,\boldsymbol{x})\,\mathrm{d}\tau\\ =&-2\mathfrak{Im}\,\mathcal{P}\int_0^t U_1(\tau,\boldsymbol{x})\,\mathrm{d}\tau = 0.\end{aligned}$$

The first equality above follows from the fact that $U_1(\tau,\boldsymbol{x})$ is a number as an operator on $L^2_{\boldsymbol{\zeta}}$.

Proposition 5.4. *The average of ρ_1 with respect to the microscopic scale vanishes,*

$$\langle\rho_1(t,\boldsymbol{x},\boldsymbol{z})\rangle_{\boldsymbol{z}} = 0, \tag{5.16}$$

which satisfies the second constraint of (4.13) *self-consistently.*

Proof. In (5.15) the first term of the right hand side is an odd function in $\boldsymbol{z}$, hence when taken the average over $\boldsymbol{z}$, it gives zero. The second term is the first order density perturbation $\widetilde{\rho}_1$ if one takes $J=1$ in Lemma 5.1, whose average over $\boldsymbol{z}$ is also zero.

□

For a more explicit expression for ρ_1, we substitute the spectral representation of operator H_0 into (5.15),

$$H_0 = \sum_n \fint_{\Gamma^*} E_{n,\boldsymbol{k}}|\psi_{n,\boldsymbol{k}}\rangle\langle\psi_{n,\boldsymbol{k}}|\,\mathrm{d}\boldsymbol{k}. \tag{5.17}$$

This gives

(5.18)
$$
\begin{aligned}
&\rho_1(t,\boldsymbol{x},\boldsymbol{z}) = \\
&\quad -2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{(\Gamma^*)^2} \psi_{n,\boldsymbol{k}}(\boldsymbol{z})\psi^*_{m,\boldsymbol{p}}(\boldsymbol{z})e^{i(E_{m,\boldsymbol{p}}-E_{n,\boldsymbol{k}})(t-\tau)} \\
&\qquad\qquad \times \int_{\mathbb{R}^3} (\boldsymbol{\zeta}-\boldsymbol{z})\psi^*_{n,\boldsymbol{k}}(\boldsymbol{\zeta})\psi_{m,\boldsymbol{p}}(\boldsymbol{\zeta})\,\mathrm{d}\boldsymbol{\zeta}\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\boldsymbol{p}\cdot\nabla_{\boldsymbol{x}}U_0(\tau,\boldsymbol{x})\,\mathrm{d}\tau \\
&\quad -2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{(\Gamma^*)^2} \psi_{n,\boldsymbol{k}}(\boldsymbol{z})\psi^*_{m,\boldsymbol{p}}(\boldsymbol{z})e^{i(E_{m,\boldsymbol{p}}-E_{n,\boldsymbol{k}})(t-\tau)} \\
&\qquad\qquad \times \int_{\mathbb{R}^3} v_1(\tau,\boldsymbol{x},\boldsymbol{\zeta})\psi^*_{n,\boldsymbol{k}}(\boldsymbol{\zeta})\psi_{m,\boldsymbol{p}}(\boldsymbol{\zeta})\,\mathrm{d}\boldsymbol{\zeta}\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\boldsymbol{p}\,\mathrm{d}\tau, \\
&\quad -2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{(\Gamma^*)^2} \psi_{n,\boldsymbol{k}}(\boldsymbol{z})\psi^*_{m,\boldsymbol{p}}(\boldsymbol{z})e^{i(E_{m,\boldsymbol{p}}-E_{n,\boldsymbol{k}})(t-\tau)} \\
&\qquad\qquad \times \int_{\mathbb{R}^3} \psi^*_{n,\boldsymbol{k}}(\boldsymbol{\zeta})(i\nabla_{\boldsymbol{\zeta}})\psi_{m,\boldsymbol{p}}(\boldsymbol{\zeta})\cdot\boldsymbol{A}_0(\tau,\boldsymbol{x})\,\mathrm{d}\boldsymbol{\zeta}\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\boldsymbol{p}\,\mathrm{d}\tau.
\end{aligned}
$$

Substituting (5.5)-(5.7) in (5.18), we obtain

$$
\begin{aligned}
\rho_1(t,\boldsymbol{x},\boldsymbol{z}) = &-2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{\Gamma^*} u_{n,\boldsymbol{k}}(\boldsymbol{z})u^*_{m,\boldsymbol{k}}(\boldsymbol{z})e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)} \\
&\qquad \times \langle u_{n,\boldsymbol{k}}|i\nabla_{\boldsymbol{k}}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\,\mathrm{d}\boldsymbol{k}\cdot\nabla_{\boldsymbol{x}}U_0(\tau,\boldsymbol{x})\,\mathrm{d}\tau \\
&-2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{\Gamma^*} u_{n,\boldsymbol{k}}(\boldsymbol{z})u^*_{m,\boldsymbol{k}}(\boldsymbol{z})e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)} \\
&\qquad \times \langle u_{n,\boldsymbol{k}}|v_1(\tau,\boldsymbol{x},\boldsymbol{\zeta})|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\tau, \\
&-2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{\Gamma^*} u_{n,\boldsymbol{k}}(\boldsymbol{z})u^*_{m,\boldsymbol{k}}(\boldsymbol{z})e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)} \\
&\qquad \times \langle u_{n,\boldsymbol{k}}|i\nabla_{\boldsymbol{\zeta}}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\cdot\boldsymbol{A}_0(\tau,\boldsymbol{x})\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\tau.
\end{aligned}
\tag{5.19}
$$

This implies (5.2).

Remark. From (5.19) and using the orthogonality of $\{u_{n,\boldsymbol{k}}\}$ for each $\boldsymbol{k}$, we once again see that

$$
\langle \rho_1(t,\boldsymbol{x},\boldsymbol{z})\rangle_{\boldsymbol{z}} = 0.
$$

5.2.2. *Derivation of the equation* (5.3). By (4.26), (4.35) and (4.30), the second order density perturbation reads as

$$\begin{aligned}\rho_2(t,\boldsymbol{x},\boldsymbol{z}) = &- 2\mathfrak{Re}\, e^{-itH_0}\mathcal{P}\int_0^t\int_0^{\tau_2} e^{i\tau_1 H_0}\delta H_1(\tau_1,\boldsymbol{x},\boldsymbol{z})e^{i(\tau_2-\tau_1)H_0}\\ &\qquad\times \delta H_1(\tau_2,\boldsymbol{x},\boldsymbol{z})e^{i(t-\tau_2)H_0}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2(\boldsymbol{z},\boldsymbol{z})\\ &- 2\mathfrak{Im}\, e^{-itH_0}\mathcal{P}\int_0^t e^{i\tau H_0}\delta H_2(\tau,\boldsymbol{x},\boldsymbol{z})e^{i(t-\tau)H_0}\,\mathrm{d}\tau(\boldsymbol{z},\boldsymbol{z})\\ &+\int_0^t e^{-i(t-\tau_1)H_0}\delta H_1(\tau_1,\boldsymbol{x},\boldsymbol{z})e^{-i\tau_1 H_0}\,\mathrm{d}\tau_1\mathcal{P}\\ &\qquad\times\int_0^t e^{i\tau_2 H_0}\delta H_1(\tau_2,\boldsymbol{x},\boldsymbol{z})e^{i(t-\tau_2)H_0}\,\mathrm{d}\tau_2(\boldsymbol{z},\boldsymbol{z}).\end{aligned}$$

Since $\mathcal{P}$ commutates with $\exp(-itH_0)$, we may simplify the above expression as

$$\begin{aligned}&\rho_2(t,\boldsymbol{x},\boldsymbol{z}) =\\ &\quad- 2\mathfrak{Re}\,\mathcal{P}\int_0^t\int_0^{\tau_2} e^{i(\tau_1-t)H_0}\delta H_1(\tau_1,\boldsymbol{x},\boldsymbol{z})e^{i(\tau_2-\tau_1)H_0}\\ &\qquad\qquad\times \delta H_1(\tau_2,\boldsymbol{x},\boldsymbol{z})e^{i(t-\tau_2)H_0}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2(\boldsymbol{z},\boldsymbol{z})\\ &\quad- 2\mathfrak{Im}\,\mathcal{P}\int_0^t e^{i(\tau-t)H_0}\delta H_2(\tau,\boldsymbol{x},\boldsymbol{z})e^{i(t-\tau)H_0}\,\mathrm{d}\tau(\boldsymbol{z},\boldsymbol{z})\\ &\quad+\int_0^t e^{-i(t-\tau_1)H_0}\delta H_1(\tau_1,\boldsymbol{x},\boldsymbol{z})\mathcal{P}\\ &\qquad\qquad\times\int_0^t e^{i(\tau_2-\tau_1)H_0}\delta H_1(\tau_2,\boldsymbol{x},\boldsymbol{z})e^{i(t-\tau_2)H_0}\,\mathrm{d}\tau_2\,\mathrm{d}\tau_1(\boldsymbol{z},\boldsymbol{z}).\end{aligned}\tag{5.20}$$

Proposition 5.5. *The average of* $\rho_2(t,\boldsymbol{x},\boldsymbol{z})$ *is given by*

$$\begin{aligned}&\langle\rho_2(t,\boldsymbol{x},\boldsymbol{z})\rangle_{\boldsymbol{z}} =\\ &\quad- 2\mathfrak{Im}\left\langle\mathcal{P}\int_0^t e^{i(\tau-t)H_0}\tfrac{1}{2}\big((\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}\big)^2U_0(\tau,\boldsymbol{x})e^{i(t-\tau)H_0}\,\mathrm{d}\tau(\boldsymbol{z},\boldsymbol{z})\right\rangle_{\boldsymbol{z}}\\ &\quad- 2\mathfrak{Im}\left\langle\mathcal{P}\int_0^t e^{i(\tau-t)H_0}(\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}v_1(\tau,\boldsymbol{x},\boldsymbol{\zeta})e^{i(t-\tau)H_0}\,\mathrm{d}\tau(\boldsymbol{z},\boldsymbol{z})\right\rangle_{\boldsymbol{z}},\\ &\quad- 2\mathfrak{Im}\left\langle\mathcal{P}\int_0^t e^{i(\tau-t)H_0}\Big((\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}\boldsymbol{A}_0(\tau,\boldsymbol{x})\Big)\cdot(i\nabla_{\boldsymbol{\zeta}})e^{i(t-\tau)H_0}\,\mathrm{d}\tau(\boldsymbol{z},\boldsymbol{z})\right\rangle_{\boldsymbol{z}}.\end{aligned}\tag{5.21}$$

Proof. Substituting the expressions of δH_1 and δH_2 (4.32)-(4.33) into (5.20) and taking average with respect to $\boldsymbol{z}$, one has

$$
\begin{aligned}
&\langle \rho_2(t,\boldsymbol{x},\boldsymbol{z})\rangle_{\boldsymbol{z}} = I_1 + I_2 \\
&\quad - 2\mathfrak{Im}\left\langle \mathcal{P}\int_0^t e^{i(\tau-t)H_0}\tfrac{1}{2}\big((\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}\big)^2 U_0(\tau,\boldsymbol{x}) e^{i(t-\tau)H_0}\,\mathrm{d}\tau(\boldsymbol{z},\boldsymbol{z})\right\rangle_{\boldsymbol{z}} \\
&\quad - 2\mathfrak{Im}\left\langle \mathcal{P}\int_0^t e^{i(\tau-t)H_0}(\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}} v_1(\tau,\boldsymbol{x},\boldsymbol{\zeta}) e^{i(t-\tau)H_0}\,\mathrm{d}\tau(\boldsymbol{z},\boldsymbol{z})\right\rangle_{\boldsymbol{z}} \\
&\quad - 2\mathfrak{Im}\left\langle \mathcal{P}\int_0^t e^{i(\tau-t)H_0}\Big((\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}\boldsymbol{A}_0(\tau,\boldsymbol{x})\Big)\cdot(i\nabla_{\boldsymbol{\zeta}}) e^{i(t-\tau)H_0}\,\mathrm{d}\tau(\boldsymbol{z},\boldsymbol{z})\right\rangle_{\boldsymbol{z}},
\end{aligned}
$$

where I_1 and I_2 are given by

$$
\begin{aligned}
I_1 = -2\mathfrak{Re}\Big\langle \mathcal{P}\int_0^t\int_0^{\tau_2} & e^{iH_0(\tau_1-t)}(\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}U_0(\tau_1)e^{iH_0(\tau_2-\tau_1)} \\
& \times(\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}U_0(\tau_2)e^{iH_0(t-\tau_2)}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2(\boldsymbol{z},\boldsymbol{z})\Big\rangle_{\boldsymbol{z}} \\
+\Big\langle \int_0^t & e^{iH_0(\tau_1-t)}(\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}U_0(\tau_1)\mathcal{P} \\
& \times\int_0^t e^{iH_0(\tau_2-\tau_1)}(\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}U_0(\tau_2)e^{iH_0(t-\tau_2)}\,\mathrm{d}\tau_2\,\mathrm{d}\tau_1(\boldsymbol{z},\boldsymbol{z})\Big\rangle_{\boldsymbol{z}},
\end{aligned}
$$

$$
\begin{aligned}
I_2 = -2\mathfrak{Re}\Big\langle \mathcal{P}\int_0^t\int_0^{\tau_2} & e^{iH_0(\tau_1-t)}(\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}U_0(\tau_1)e^{iH_0(\tau_2-\tau_1)} \\
& \times\Big(v_1(\tau_2)+\boldsymbol{A}_0(\tau_2)\cdot(i\nabla_{\boldsymbol{\zeta}})\Big)e^{iH_0(t-\tau_2)}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2(\boldsymbol{z},\boldsymbol{z})\Big\rangle_{\boldsymbol{z}} \\
-2\mathfrak{Re}\Big\langle \mathcal{P}\int_0^t\int_0^{\tau_2} & e^{iH_0(\tau_1-t)}\Big(v_1(\tau_1)+\boldsymbol{A}_0(\tau_1)\cdot(i\nabla_{\boldsymbol{\zeta}})\Big)e^{iH_0(\tau_2-\tau_1)} \\
& \times(\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}U_0(\tau_2)e^{iH_0(t-\tau_2)}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2(\boldsymbol{z},\boldsymbol{z})\Big\rangle_{\boldsymbol{z}} \\
+2\mathfrak{Re}\Big\langle \int_0^t & e^{iH_0(\tau_1-t)}(\boldsymbol{\zeta}-\boldsymbol{z})\cdot\nabla_{\boldsymbol{x}}U_0(\tau_1)\mathcal{P} \\
& \times\int_0^t e^{iH_0(\tau_2-\tau_1)}\Big(v_1(\tau_2)+\boldsymbol{A}_0(\tau_2)\cdot(i\nabla_{\boldsymbol{\zeta}})\Big)e^{iH_0(t-\tau_2)}\,\mathrm{d}\tau_2\,\mathrm{d}\tau_1(\boldsymbol{z},\boldsymbol{z})\Big\rangle_{\boldsymbol{z}}.
\end{aligned}
$$

Remark that when calculating $\langle \rho_2(t,\boldsymbol{x},\boldsymbol{z})\rangle_{\boldsymbol{z}}$, we have dropped out the $\boldsymbol{z}$-average of the odd functions in $\boldsymbol{z}$ and the second order density perturbation functions of $\widetilde{H} = H_0 + \varepsilon(v_1 + U_1 + i\boldsymbol{A}_0\cdot\nabla_{\boldsymbol{\zeta}}) + \varepsilon^2(V_2 + \frac{1}{2}|\boldsymbol{A}_0|^2 + i\boldsymbol{A}_1\cdot\nabla_{\boldsymbol{\zeta}})$ by Lemma 5.1.

We complete the proof by showing that $I_1 = 0$ and $I_2 = 0$.

We denote the second term in I_1 as $I_{1,2} = \mathfrak{Re}\,\langle \int_0^t \int_0^t K(\tau_1,\tau_2)\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2(\boldsymbol{z},\boldsymbol{z})\rangle_{\boldsymbol{z}}$, then

$$\begin{aligned} I_{1,2} &= \mathfrak{Re}\left\langle \int_0^t \int_0^{\tau_1} K(\tau_1,\tau_2)\,\mathrm{d}\tau_2\,\mathrm{d}\tau_1(\boldsymbol{z},\boldsymbol{z})\right\rangle_{\boldsymbol{z}} \\ &\quad + \mathfrak{Re}\left\langle \int_0^t \int_0^{\tau_2} K(\tau_1,\tau_2)\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2(\boldsymbol{z},\boldsymbol{z})\right\rangle_{\boldsymbol{z}} \\ &= 2\mathfrak{Re}\left\langle \int_0^t \int_0^{\tau_2} K(\tau_1,\tau_2)\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2(\boldsymbol{z},\boldsymbol{z})\right\rangle_{\boldsymbol{z}}, \end{aligned}$$

where the last equality is obtained by switching $\tau_1 \leftrightarrow \tau_2$ in the first term of $I_{1,2}$ and using the fact that $K(\tau_1,\tau_2) = \overline{K(\tau_2,\tau_1)}$. Therefore I_1 could be rewritten as

$$\begin{aligned} I_1 = -2\mathfrak{Re}&\left\langle \mathcal{P}\int_0^t \int_0^{\tau_2} e^{iH_0(\tau_1-t)}(\boldsymbol{\zeta}-\boldsymbol{z})_\alpha e^{iH_0(\tau_2-\tau_1)} \right. \\ &\qquad \left. \times (\boldsymbol{\zeta}-\boldsymbol{z})_\beta e^{iH_0(t-\tau_2)} U^{\alpha\beta}_{\tau_1\tau_2}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2(\boldsymbol{z},\boldsymbol{z})\right\rangle_{\boldsymbol{z}} \\ +2\mathfrak{Re}&\left\langle \int_0^t \int_0^{\tau_2} e^{iH_0(\tau_1-t)}(\boldsymbol{\zeta}-\boldsymbol{z})_\alpha \mathcal{P} e^{iH_0(\tau_2-\tau_1)} \right. \\ &\qquad \left. \times (\boldsymbol{\zeta}-\boldsymbol{z})_\beta e^{iH_0(t-\tau_2)} U^{\alpha\beta}_{\tau_1\tau_2}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2(\boldsymbol{z},\boldsymbol{z})\right\rangle_{\boldsymbol{z}}, \end{aligned} \tag{5.22}$$

where we have used the short hand notation $U^{\alpha\beta}_{\tau_1\tau_2} = \partial_{\boldsymbol{x}_\alpha} U_0(\tau_1)\partial_{\boldsymbol{x}_\beta} U_0(\tau_2)$.

Substituting spectral representation of H_0 (5.17) into (5.22) gives

$$\begin{aligned} I_1 = -2\mathfrak{Re}\sum_{n\le Z}\sum_{m\ell}&\left\langle \int_0^t \int_0^{\tau_2} \fint_{(\Gamma^*)^3} \psi_{n,\boldsymbol{k}}(\boldsymbol{z})\psi^*_{\ell,\boldsymbol{q}}(\boldsymbol{z}) e^{iE_{n,\boldsymbol{k}}(\tau_1-t)} F^\alpha_{n,\boldsymbol{k};m,\boldsymbol{p}} \right. \\ &\left. \times e^{iE_{m,\boldsymbol{p}}(\tau_2-\tau_1)} F^\beta_{m,\boldsymbol{p};\ell,\boldsymbol{q}} e^{iE_{\ell,\boldsymbol{q}}(t-\tau_2)} U^{\alpha\beta}_{\tau_1\tau_2}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\boldsymbol{p}\,\mathrm{d}\boldsymbol{q}\right\rangle_{\boldsymbol{z}} \\ +2\mathfrak{Re}\sum_{n\le Z}\sum_{m\ell}&\left\langle \int_0^t \int_0^{\tau_2} \fint_{(\Gamma^*)^3} \psi_{m,\boldsymbol{p}}(\boldsymbol{z})\psi^*_{\ell,\boldsymbol{q}}(\boldsymbol{z}) e^{iE_{m,\boldsymbol{p}}(\tau_1-t)} F^\alpha_{m,\boldsymbol{p};n,\boldsymbol{k}} \right. \\ &\left. \times e^{iE_{n,\boldsymbol{k}}(\tau_2-\tau_1)} F^\beta_{n,\boldsymbol{k};\ell,\boldsymbol{q}} e^{iE_{\ell,\boldsymbol{q}}(t-\tau_2)} U^{\alpha\beta}_{\tau_1\tau_2}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\boldsymbol{p}\,\mathrm{d}\boldsymbol{q}\right\rangle_{\boldsymbol{z}}, \end{aligned}$$

where $F^\alpha_{n,\boldsymbol{k};m,\boldsymbol{p}} = \int_{\mathbb{R}^3}(\boldsymbol{\zeta}-\boldsymbol{z})_\alpha \psi^*_{n,\boldsymbol{k}}(\boldsymbol{\zeta})\psi_{m,\boldsymbol{p}}(\boldsymbol{\zeta})\,\mathrm{d}\boldsymbol{\zeta}$, and $F^\alpha_{m,\boldsymbol{p};n,\boldsymbol{k}}$, $F^\beta_{m,\boldsymbol{p};\ell,\boldsymbol{q}}$, $F^\beta_{n,\boldsymbol{k};\ell,\boldsymbol{q}}$ are defined similarly.

We denote I_1 as

$$I_1 = -2\mathfrak{Re}\sum_{n\le Z}\sum_{m\ell} N^{nml}_{\boldsymbol{kpq}} + 2\mathfrak{Re}\sum_{n\le Z}\sum_{m\ell} N^{mnl}_{\boldsymbol{pkq}},$$

then it is easy to see that, by switching $(m,\boldsymbol{p})$ and $(n,\boldsymbol{k})$,

$$-\mathfrak{Re}\sum_{n\le Z}\sum_{m\le Z}\sum_{\ell} N^{nml}_{\boldsymbol{kpq}} + \mathfrak{Re}\sum_{n\le Z}\sum_{m\le Z}\sum_{\ell} N^{mnl}_{\boldsymbol{pkq}} = 0.$$

Therefore

$$I_1 = -2\mathfrak{Re} \sum_{n\le Z}\sum_{m>Z}\sum_{\ell} N^{nml}_{\boldsymbol{kpq}} + 2\mathfrak{Re} \sum_{n\le Z}\sum_{m>Z}\sum_{\ell} N^{mnl}_{\boldsymbol{pkq}}.$$

Making use of the identity (5.5) produces

$$\begin{aligned} F^{\alpha}_{n,\boldsymbol{k};m,\boldsymbol{p}} &= \Big\{\delta_{nm}(-\boldsymbol{z}_\alpha - i\partial_{\boldsymbol{p}_\alpha}) + \langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\Big\}\delta(\boldsymbol{p}-\boldsymbol{k})\,|\Gamma^*|\,, \\ F^{\beta}_{m,\boldsymbol{p};\ell,\boldsymbol{q}} &= \Big\{\delta_{m\ell}(-\boldsymbol{z}_\beta - i\partial_{\boldsymbol{q}_\beta}) + \langle u_{m,\boldsymbol{p}}|i\partial_{\boldsymbol{p}_\beta}|u_{\ell,\boldsymbol{p}}\rangle_{L^2(\Gamma)}\Big\}\delta(\boldsymbol{q}-\boldsymbol{p})\,|\Gamma^*|\,. \end{aligned}$$

Then by the orthogonality of $\{u_{n,\boldsymbol{k}}\}$ for each $\boldsymbol{k}$ and using integration by parts for the variable $\boldsymbol{p}$, one could rewrite $I_1 = I_1^{(1)} + I_1^{(2)}$ which are given by

$$\begin{aligned} I_1^{(1)} = \\ & - 2\mathfrak{Re} \sum_{n\le Z}\sum_{m>Z}\sum_{\ell} \Big\langle \int_0^t\int_0^{\tau_2} \fint_{(\Gamma^*)^2} \psi_{n,\boldsymbol{k}}\psi^*_{\ell,\boldsymbol{q}} e^{iE_{n,\boldsymbol{k}}(\tau_1-t)}\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \\ & \qquad \times e^{iE_{m,\boldsymbol{k}}(\tau_2-\tau_1)}\delta_{m\ell}(-\boldsymbol{z}_\beta - i\partial_{\boldsymbol{q}_\beta})\delta(\boldsymbol{q}-\boldsymbol{k})e^{iE_{\ell,\boldsymbol{q}}(t-\tau_2)}U^{\alpha\beta}_{\tau_1\tau_2}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\boldsymbol{q}\Big\rangle_{\boldsymbol{z}} \\ & + 2\mathfrak{Re} \sum_{n\le Z}\sum_{m>Z}\sum_{\ell} \Big\langle \int_0^t\int_0^{\tau_2} \fint_{(\Gamma^*)^2} \psi_{m,\boldsymbol{k}}\psi^*_{\ell,\boldsymbol{q}} e^{iE_{m,\boldsymbol{k}}(\tau_1-t)}\langle u_{m,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{n,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \\ & \qquad \times e^{iE_{n,\boldsymbol{k}}(\tau_2-\tau_1)}\delta_{n\ell}(-\boldsymbol{z}_\beta - i\partial_{\boldsymbol{q}_\beta})\delta(\boldsymbol{q}-\boldsymbol{k})e^{iE_{\ell,\boldsymbol{q}}(t-\tau_2)}U^{\alpha\beta}_{\tau_1\tau_2}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\boldsymbol{q}\Big\rangle_{\boldsymbol{z}} \\ = & + 2\mathfrak{Re} \sum_{n\le Z}\sum_{m>Z} \Big\langle \int_0^t\int_0^{\tau_2} \fint_{\Gamma^*} \langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)} \\ & \qquad \times e^{i(E_{m,\boldsymbol{k}}-E_{n,\boldsymbol{k}})(t-\tau_1)}U^{\alpha\beta}_{\tau_1\tau_2}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2\,\mathrm{d}\boldsymbol{k}\Big\rangle_{\boldsymbol{z}} \\ & - 2\mathfrak{Re} \sum_{n\le Z}\sum_{m>Z} \Big\langle \int_0^t\int_0^{\tau_2} \fint_{\Gamma^*} \langle u_{m,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{n,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\overline{\langle u_{m,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{n,\boldsymbol{k}}\rangle}_{L^2(\Gamma)} \\ & \qquad e^{i(E_{n,\boldsymbol{k}}-E_{m,\boldsymbol{k}})(t-\tau_1)}U^{\alpha\beta}_{\tau_1\tau_2}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2\,\mathrm{d}\boldsymbol{k}\Big\rangle_{\boldsymbol{z}}\,, \end{aligned}$$

$$
\begin{aligned}
I_1^{(2)} = &-2\mathfrak{Re} \sum_{n\le Z}\sum_{m>Z}\sum_{\ell}\Big\langle \int_0^t\int_0^{\tau_2} \fint_{\Gamma^*} \psi_{n,\boldsymbol{k}}\psi^*_{\ell,\boldsymbol{k}} e^{iE_{n,\boldsymbol{k}}(\tau_1-t)}\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \\
&\times e^{iE_{m,\boldsymbol{k}}(\tau_2-\tau_1)}\langle u_{m,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{\ell,\boldsymbol{k}}\rangle_{L^2(\Gamma)} e^{iE_{\ell,\boldsymbol{k}}(t-\tau_2)} U^{\alpha\beta}_{\tau_1\tau_2}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2\,\mathrm{d}\boldsymbol{k}\Big\rangle_{\boldsymbol{z}} \\
&+2\mathfrak{Re} \sum_{n\le Z}\sum_{m>Z}\sum_{\ell}\Big\langle \int_0^t\int_0^{\tau_2} \fint_{\Gamma^*} \psi_{m,\boldsymbol{k}}\psi^*_{\ell,\boldsymbol{k}} e^{iE_{m,\boldsymbol{k}}(\tau_1-t)}\langle u_{m,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{n,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \\
&\times e^{iE_{n,\boldsymbol{k}}(\tau_2-\tau_1)}\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{\ell,\boldsymbol{k}}\rangle_{L^2(\Gamma)} e^{iE_{\ell,\boldsymbol{k}}(t-\tau_2)} U^{\alpha\beta}_{\tau_1\tau_2}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2\,\mathrm{d}\boldsymbol{k}\Big\rangle_{\boldsymbol{z}} \\
= &-2\mathfrak{Re} \sum_{n\le Z}\sum_{m>Z}\Big\langle \int_0^t\int_0^{\tau_2} \fint_{\Gamma^*} \langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\langle u_{m,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{n,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \\
&\times e^{i(E_{m,\boldsymbol{k}}-E_{n,\boldsymbol{k}})(\tau_2-\tau_1)} U^{\alpha\beta}_{\tau_1\tau_2}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2\,\mathrm{d}\boldsymbol{k}\Big\rangle_{\boldsymbol{z}} \\
&+2\mathfrak{Re} \sum_{n\le Z}\sum_{m>Z}\Big\langle \int_0^t\int_0^{\tau_2} \fint_{\Gamma^*} \langle u_{m,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{n,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \\
&e^{i(E_{n,\boldsymbol{k}}-E_{m,\boldsymbol{k}})(\tau_2-\tau_1)} U^{\alpha\beta}_{\tau_1\tau_2}\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2\,\mathrm{d}\boldsymbol{k}\Big\rangle_{\boldsymbol{z}}.
\end{aligned}
$$

By observing that

$$
\overline{\langle u_{m,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{n,\boldsymbol{k}}\rangle}_{L^2(\Gamma)} = \langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)},
$$

we get $I_1^{(1)} = 0$ and $I_1^{(2)} = 0$ since one has the same real part as its complex conjugate.

Therefore $I_1 = 0$. Similar arguments will show that $I_2 = 0$ by making use of the identity (5.6), and we omit its details here. □

Substituting the spectral representation of H_0 (5.17) into (5.21) gives

$$
\begin{aligned}
&\langle \rho_2(t,\boldsymbol{x},\boldsymbol{z})\rangle_{\boldsymbol{z}} = \\
&\quad -2\mathfrak{Im}\Big\langle \sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{(\Gamma^*)^2} \psi_{n,\boldsymbol{k}}(\boldsymbol{z})\psi^*_{m,\boldsymbol{p}}(\boldsymbol{z}) e^{i(E_{m,\boldsymbol{p}}-E_{n,\boldsymbol{k}})(t-\tau)} \\
&\qquad \times \frac{1}{2}\int_{\mathbb{R}^3} (\boldsymbol{\zeta}-\boldsymbol{z})_\alpha(\boldsymbol{\zeta}-\boldsymbol{z})_\beta \psi^*_{n,\boldsymbol{k}}(\boldsymbol{\zeta})\psi_{m,\boldsymbol{p}}\,\mathrm{d}\boldsymbol{\zeta}\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\boldsymbol{p}\partial_{\boldsymbol{x}_\alpha}\partial_{\boldsymbol{x}_\beta}U_0(\tau,\boldsymbol{x})\,\mathrm{d}\tau\Big\rangle_{\boldsymbol{z}} \\
&\quad -2\mathfrak{Im}\Big\langle \sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{(\Gamma^*)^2} \psi_{n,\boldsymbol{k}}(\boldsymbol{z})\psi^*_{m,\boldsymbol{p}}(\boldsymbol{z}) e^{i(E_{m,\boldsymbol{p}}-E_{n,\boldsymbol{k}})(t-\tau)} \\
&\qquad \times \int_{\mathbb{R}^3} (\boldsymbol{\zeta}-\boldsymbol{z})_\alpha \partial_{\boldsymbol{x}_\alpha} v_1(\tau,\boldsymbol{x},\boldsymbol{\zeta})\psi^*_{n,\boldsymbol{k}}(\boldsymbol{\zeta})\psi_{m,\boldsymbol{p}}\,\mathrm{d}\boldsymbol{\zeta}\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\boldsymbol{p}\,\mathrm{d}\tau\Big\rangle_{\boldsymbol{z}}.
\end{aligned}
$$

By (5.11) and (5.12) and using the integration by parts with respect to $\boldsymbol{p}$, we could simplify the above equality to be

$$
\begin{aligned}
&\langle \rho_2(t,\boldsymbol{x},\boldsymbol{z})\rangle_{\boldsymbol{z}} = \\
&\quad 2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{\Gamma^*} e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)} \\
&\qquad \times \overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)}\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\partial_{\boldsymbol{x}_\alpha}\partial_{\boldsymbol{x}_\beta}U_0(\tau)\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\tau \\
&\quad + 2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{\Gamma^*} e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)} \\
&\qquad \times \overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)}\langle u_{n,\boldsymbol{k}}|\partial_{\boldsymbol{x}_\alpha}v_1(\tau)|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\tau, \\
&\quad + 2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{\Gamma^*} e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)} \\
&\qquad \times \overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)}\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{\zeta}_\beta}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\partial_{\boldsymbol{x}_\alpha}(\boldsymbol{A}_0)_\beta\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\tau.
\end{aligned}
$$

Therefore taking the $\boldsymbol{z}$-average of (4.7) produces

$$
\begin{aligned}
-\delta_{\alpha\beta}\partial_{\boldsymbol{x}_\alpha}\partial_{\boldsymbol{x}_\beta}U_0 &= \langle \rho_2(t,\boldsymbol{x},\boldsymbol{z})\rangle_{\boldsymbol{z}} + \rho_{\text{ext}}(t,\boldsymbol{x}) \\
&= P_{\alpha\beta}(\partial_{\boldsymbol{x}_\alpha}\partial_{\boldsymbol{x}_\beta}U_0) + Q_\alpha(\partial_{\boldsymbol{x}_\alpha}v_1) + R_{\alpha\beta}(\partial_{\boldsymbol{x}_\alpha(\boldsymbol{A}_0)_\beta} + \rho_{\text{ext}}(t,\boldsymbol{x}).
\end{aligned}
$$

This proves (5.3).

5.2.3. *Derivation of the equation* (5.4). Substituting the spectral representation of H_0 (5.17) into (4.27) yields

$$
\boldsymbol{J}_0 = \sum_{n\le Z}\fint_{\Gamma^*}\mathfrak{Im}\,\psi_{n,\boldsymbol{k}}\nabla_{\boldsymbol{\zeta}}\psi_{n,\boldsymbol{k}}\,\mathrm{d}\boldsymbol{k}. \tag{5.23}
$$

Proposition 5.6. *We have*

$$
\langle \boldsymbol{J}_0\rangle_{\boldsymbol{z}} = 0,
$$

which satisfies the third constraint in (4.13) *self-consistently.*

Proof. By definition of the Bloch decomposition, we have

$$
\widetilde{H_0}u_{n,\boldsymbol{k}} = \Big(\frac{1}{2}(-i\nabla_{\boldsymbol{z}}+\boldsymbol{k})^2 + v_0(\boldsymbol{z})\Big)u_{n,\boldsymbol{k}}(\boldsymbol{z}) = E_{n,\boldsymbol{k}}u_{n,\boldsymbol{k}}(\boldsymbol{z}). \tag{5.24}
$$

Differentiating (5.24) with respect to $\boldsymbol{k}$ gives

$$
(-i\nabla_{\boldsymbol{\zeta}}+\boldsymbol{k})u_{n,\boldsymbol{k}} + \widetilde{H_0}\nabla_{\boldsymbol{k}}u_{n,\boldsymbol{k}} = \nabla_{\boldsymbol{k}}E_{n,\boldsymbol{k}}u_{n,\boldsymbol{k}} + E_{n,\boldsymbol{k}}\nabla_{\boldsymbol{k}}u_{n,\boldsymbol{k}}.
$$

Since $\widetilde{H_0}$ is a self-adjoint operator, the above equation taken the inner product with $u_{n,\boldsymbol{k}}$ yields

$$
\langle u_{n,\boldsymbol{k}}|-i\nabla_{\boldsymbol{\zeta}}+\boldsymbol{k}|u_{n,\boldsymbol{k}}\rangle_{L^2(\Gamma)} = \nabla_{\boldsymbol{k}}E_{n,\boldsymbol{k}}.
$$

Therefore by (5.23),

$$
\langle \boldsymbol{J}_0\rangle_{\boldsymbol{z}} = \sum_{n\le Z}\fint_{\Gamma^*}\langle u_{n,\boldsymbol{k}}|-i\nabla_{\boldsymbol{\zeta}}+\boldsymbol{k}|u_{n,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\,\mathrm{d}\boldsymbol{k} = \sum_{n\le Z}\fint_{\Gamma^*}\nabla_{\boldsymbol{k}}E_{n,\boldsymbol{k}}\,\mathrm{d}\boldsymbol{k} = 0,
$$

where the last equality is due to the periodicity. □

Similar to the derivation of (5.19), by making use of Lemma 5.2 and (4.13), direct calculations from (4.28) give the following proposition.

Proposition 5.7. *The average of* $\boldsymbol{J}_1(t,\boldsymbol{x},\boldsymbol{z})$ *is given by*

$$
\begin{aligned}
\langle \boldsymbol{J}_1\rangle_{\boldsymbol{z}} =& 2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\int_0^t \fint_{\Gamma^*} \overline{\langle u_{n,\boldsymbol{k}}|i\nabla_{\boldsymbol{\zeta}}|u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)} e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)} \\
&\times\Big(\langle u_{n,\boldsymbol{k}}|i\nabla_{\boldsymbol{k}}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\cdot\nabla_{\boldsymbol{x}}U_0(\tau)+\langle u_{n,\boldsymbol{k}}|v_1(\tau)|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \\
&+\langle u_{m,\boldsymbol{k}}|i\nabla_{\boldsymbol{\zeta}}|u_{n,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\cdot\boldsymbol{A}_0(\tau)\Big)\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\tau-\boldsymbol{A}_0\langle\rho_0\rangle_{\boldsymbol{z}}.
\end{aligned}
\tag{5.25}
$$

Then (5.25) implies (5.4) by taking the $\boldsymbol{z}$-average of (4.12).

6. Effective equations in the frequency domain

We now derive the effective equations in frequency domain. We start with the following proposition of the Fourier transform.

Proposition 6.1. *Define the function* $h(t)=\int_0^t h_1(t-\tau)\cdot h_2(\tau)\,\mathrm{d}\tau$*, then*

$$\widehat{h}=\widehat{h_1H_v}\cdot\widehat{h_2H_v},$$

where $H_v(t)$ *is the Heaviside function of* t*,*

$$H_v(t)=\begin{cases}1 & t\ge 0,\\ 0 & \textit{otherwise}.\end{cases}$$

Without loss of generality, we assume that $U_0(t)$ and $\boldsymbol{A}_0(t)$ vanish for $t<0$. By taking the Fourier transform of (5.1)-(5.4) and using Proposition 6.1, we have

$$-\big(\delta_{\alpha\beta}+A_{\alpha\beta}\big)\partial_{\boldsymbol{x}_\alpha}\partial_{\boldsymbol{x}_\beta}\widehat{U_0}=B_{\alpha\beta}\big(\partial_{\boldsymbol{x}_\alpha}(\widehat{\boldsymbol{A}_0})_\beta\big)+\widehat{\rho_{\mathrm{ext}}},\tag{6.1}$$

$$-\omega^2(\widehat{\boldsymbol{A}_0})_\alpha-\Delta_{\boldsymbol{x}}(\widehat{\boldsymbol{A}_0})_\alpha+(-i\omega)\partial_{\boldsymbol{x}_\alpha}\widehat{U_0}=C_{\alpha\beta}\partial_{\boldsymbol{x}_\beta}\widehat{U_0}+D_{\alpha\beta}(\widehat{\boldsymbol{A}_0})_\beta+\widehat{\boldsymbol{J}_{\mathrm{ext}}}.\tag{6.2}$$

The coefficients are given by

$$
\begin{aligned}
A_{\alpha\beta}&=\widehat{P}_{\alpha\beta}-\langle\widehat{\boldsymbol{f}}_\alpha^*\mathcal{V}(I-\widehat{\chi}_\omega\mathcal{V})^{-1}\widehat{\boldsymbol{f}}_\beta\rangle_{\boldsymbol{z}},\\
B_{\alpha\beta}&=\widehat{R}_{\alpha\beta}-\langle\widehat{\boldsymbol{f}}_\alpha^*\mathcal{V}(I-\widehat{\chi}_\omega\mathcal{V})^{-1}\widehat{\boldsymbol{g}}_\beta\rangle_{\boldsymbol{z}},\\
C_{\alpha\beta}&=\widehat{M}_{\alpha\beta}-\langle\widehat{\boldsymbol{g}}_\alpha^*\mathcal{V}(I-\widehat{\chi}_\omega\mathcal{V})^{-1}\widehat{\boldsymbol{f}}_\beta\rangle_{\boldsymbol{z}},\\
D_{\alpha\beta}&=\widehat{N}_{\alpha\beta}-\langle\widehat{\boldsymbol{g}}_\alpha^*\mathcal{V}(I-\widehat{\chi}_\omega\mathcal{V})^{-1}\widehat{\boldsymbol{g}}_\beta\rangle_{\boldsymbol{z}}-\delta_{\alpha\beta}\langle\rho_0\rangle_{\boldsymbol{z}},
\end{aligned}
$$

where

$$
\begin{aligned}
\widehat{\chi}_\omega \widehat{v_1}(\omega) = &- \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega+\omega_{mn}(\boldsymbol{k})} u_{n,\boldsymbol{k}} u_{m,\boldsymbol{k}}^* \langle u_{n,\boldsymbol{k}} | \widehat{v_1}(\omega) | u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \,\mathrm{d}\boldsymbol{k} \\
&+ \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega-\omega_{mn}(\boldsymbol{k})} u_{n,\boldsymbol{k}}^* u_{m,\boldsymbol{k}} \overline{\langle u_{n,\boldsymbol{k}} | \widehat{v_1}(\omega) | u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)} \,\mathrm{d}\boldsymbol{k}, \\
\widehat{\boldsymbol{f}}(\omega) = &- \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega+\omega_{mn}(\boldsymbol{k})} u_{n,\boldsymbol{k}} u_{m,\boldsymbol{k}}^* \langle u_{n,\boldsymbol{k}} | i\nabla_{\boldsymbol{k}} | u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \,\mathrm{d}\boldsymbol{k} \\
&+ \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega-\omega_{mn}(\boldsymbol{k})} u_{n,\boldsymbol{k}}^* u_{m,\boldsymbol{k}} \overline{\langle u_{n,\boldsymbol{k}} | i\nabla_{\boldsymbol{k}} | u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)} \,\mathrm{d}\boldsymbol{k}, \\
\widehat{\boldsymbol{g}}(\omega) = &- \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega+\omega_{mn}(\boldsymbol{k})} u_{n,\boldsymbol{k}} u_{m,\boldsymbol{k}}^* \langle u_{n,\boldsymbol{k}} | i\nabla_{\boldsymbol{\zeta}} | u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)} \,\mathrm{d}\boldsymbol{k} \\
&+ \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega-\omega_{mn}(\boldsymbol{k})} u_{n,\boldsymbol{k}}^* u_{m,\boldsymbol{k}} \overline{\langle u_{n,\boldsymbol{k}} | i\nabla_{\boldsymbol{\zeta}} | u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)} \,\mathrm{d}\boldsymbol{k},
\end{aligned}
$$

and

$$
\begin{aligned}
\widehat{P}_{\alpha\beta}(\omega) = &\sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega+\omega_{mn}(\boldsymbol{k})} \overline{\langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{k}_\alpha} | u_{m,\boldsymbol{k}}\rangle} \langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{k}_\beta} | u_{m,\boldsymbol{k}}\rangle \,\mathrm{d}\boldsymbol{k} \\
&- \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega-\omega_{mn}(\boldsymbol{k})} \langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{k}_\alpha} | u_{m,\boldsymbol{k}}\rangle \overline{\langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{k}_\beta} | u_{m,\boldsymbol{k}}\rangle} \,\mathrm{d}\boldsymbol{k},
\end{aligned}
$$

$$
\begin{aligned}
\widehat{R}_{\alpha\beta}(\omega) = &\sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega+\omega_{mn}(\boldsymbol{k})} \overline{\langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{k}_\alpha} | u_{m,\boldsymbol{k}}\rangle} \langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{\zeta}_\beta} | u_{m,\boldsymbol{k}}\rangle \,\mathrm{d}\boldsymbol{k} \\
&- \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega-\omega_{mn}(\boldsymbol{k})} \langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{k}_\alpha} | u_{m,\boldsymbol{k}}\rangle \overline{\langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{\zeta}_\beta} | u_{m,\boldsymbol{k}}\rangle} \,\mathrm{d}\boldsymbol{k},
\end{aligned}
$$

$$
\begin{aligned}
\widehat{M}_{\alpha\beta}(\omega) = &\sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega+\omega_{mn}(\boldsymbol{k})} \overline{\langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{\zeta}_\alpha} | u_{m,\boldsymbol{k}}\rangle} \langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{k}_\beta} | u_{m,\boldsymbol{k}}\rangle \,\mathrm{d}\boldsymbol{k} \\
&- \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega-\omega_{mn}(\boldsymbol{k})} \langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{\zeta}_\alpha} | u_{m,\boldsymbol{k}}\rangle \overline{\langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{k}_\beta} | u_{m,\boldsymbol{k}}\rangle} \,\mathrm{d}\boldsymbol{k},
\end{aligned}
$$

$$
\begin{aligned}
\widehat{N}_{\alpha\beta}(\omega) = &\sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega+\omega_{mn}(\boldsymbol{k})} \overline{\langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{\zeta}_\alpha} | u_{m,\boldsymbol{k}}\rangle} \langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{\zeta}_\beta} | u_{m,\boldsymbol{k}}\rangle \,\mathrm{d}\boldsymbol{k} \\
&- \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \frac{1}{\omega-\omega_{mn}(\boldsymbol{k})} \langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{\zeta}_\alpha} | u_{m,\boldsymbol{k}}\rangle \overline{\langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{\zeta}_\beta} | u_{m,\boldsymbol{k}}\rangle} \,\mathrm{d}\boldsymbol{k}.
\end{aligned}
$$

We need the following proposition to further simplify the equations.

Proposition 6.2.

$$\langle u_{n,\boldsymbol{k}} | i\nabla_{\boldsymbol{\zeta}} | u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)} = i\omega_{mn}(\boldsymbol{k}) \langle u_{n,\boldsymbol{k}} | i\nabla_{\boldsymbol{k}} | u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}.$$

Proof. Similar to (5.24) one has

$$\widetilde{H}_0 u_{m,\boldsymbol{k}} = \left(\frac{1}{2}(-i\nabla_{\boldsymbol{z}}+\boldsymbol{k})^2 + v_0(\boldsymbol{z})\right)u_{m,\boldsymbol{k}}(\boldsymbol{z}) = E_{m,\boldsymbol{k}}u_{m,\boldsymbol{k}}(\boldsymbol{z}).$$

Differentiating it with respect to $\boldsymbol{k}$ gives

$$(-i\nabla_{\boldsymbol{\zeta}}+\boldsymbol{k})u_{m,\boldsymbol{k}} + \widetilde{H}_0\nabla_{\boldsymbol{k}}u_{m,\boldsymbol{k}} = \nabla_{\boldsymbol{k}}E_{m,\boldsymbol{k}}u_{m,\boldsymbol{k}} + E_{m,\boldsymbol{k}}\nabla_{\boldsymbol{k}}u_{m,\boldsymbol{k}}.$$

Since $\widetilde{H}_0$ is a self-adjoint operator, the above equation taken the inner product with $u_{n,\boldsymbol{k}}$ produces

$$\langle u_{n,\boldsymbol{k}}|-i\nabla_{\boldsymbol{\zeta}}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)} = (E_{m,\boldsymbol{k}}-E_{n,\boldsymbol{k}})\langle u_{n,\boldsymbol{k}}|\nabla_{\boldsymbol{k}}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)},$$

which implies the conclusion. □

Lemma 6.3.

$$2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\fint_{\Gamma^*} u_{n,\boldsymbol{k}}(\boldsymbol{z})u^*_{m,\boldsymbol{k}}(\boldsymbol{z})\langle u_{n,\boldsymbol{k}}|i\nabla_{\boldsymbol{k}}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\,\mathrm{d}\boldsymbol{k} = 0, \tag{6.3}$$

$$\begin{aligned}&-2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\fint_{\Gamma^*}\overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{\zeta}_\alpha}|u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)}\\ &\qquad\qquad\times\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\,\mathrm{d}\boldsymbol{k} = \langle\rho_0\rangle_{\boldsymbol{z}}\delta_{\alpha\beta}.\end{aligned} \tag{6.4}$$

Proof. Since adding any constant vector to $\boldsymbol{A}_0$ will not change the system (3.18)-(3.20), the values of ρ_1 and $\langle\boldsymbol{J}_1\rangle_{\boldsymbol{z}}$ remain the same under the transform $\boldsymbol{A}_0 \to \boldsymbol{A}_0 + \boldsymbol{C}_{\boldsymbol{v}}$ where $\boldsymbol{C}_{\boldsymbol{v}}$ is an arbitrary constant vector.

Note that we have assumed $\boldsymbol{A}_0(t) = 0$ for $t<0$, then (5.19) implies that

$$\begin{aligned}2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\int_{-\infty}^{t}\fint_{\Gamma^*} u_{n,\boldsymbol{k}}(\boldsymbol{z})u^*_{m,\boldsymbol{k}}(\boldsymbol{z})e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)}&\\ \times\langle u_{n,\boldsymbol{k}}|i\nabla_{\boldsymbol{k}}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\tau &= 0,\end{aligned}$$

which gives (6.3) by making use of Proposition 6.2.

Similarly (5.25) implies

$$\begin{aligned}&-2\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\int_{-\infty}^{t}\fint_{\Gamma^*} e^{i\omega_{mn}(\boldsymbol{k})(t-\tau)}\\ &\quad\times\overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{\zeta}_\alpha}|u_{m,\boldsymbol{k}}\rangle}_{L^2(\Gamma)}\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{m,\boldsymbol{k}}\rangle_{L^2(\Gamma)}\,\mathrm{d}\boldsymbol{k}\,\mathrm{d}\tau = \langle\rho_0\rangle_{\boldsymbol{z}}\delta_{\alpha\beta},\end{aligned}$$

which produces (6.4) by making use of Proposition 6.2. □

Lemma 6.4.

$$\widehat{\boldsymbol{g}}(\omega) = (-i\omega)\widehat{\boldsymbol{f}}(\omega),\quad \widehat{N}_{\alpha\beta}(\omega) - \langle\rho_0\rangle_{\boldsymbol{z}}\delta_{\alpha\beta} = (-i\omega)\widehat{M}_{\alpha\beta}(\omega),$$
$$\widehat{M}_{\alpha\beta}(\omega) = -\widehat{R}_{\alpha\beta}(\omega),\quad \widehat{R}_{\alpha\beta}(\omega) = (-i\omega)\big(\widehat{P}_{\alpha\beta}(\omega) - P^r_{\alpha\beta}\big),$$

where

$$P^r_{\alpha\beta} = \frac{2i}{\omega}\mathfrak{Im}\sum_{n\le Z}\sum_{m>Z}\fint_{\Gamma^*}\overline{\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\alpha}|u_{m,\boldsymbol{k}}\rangle}\langle u_{n,\boldsymbol{k}}|i\partial_{\boldsymbol{k}_\beta}|u_{m,\boldsymbol{k}}\rangle\,\mathrm{d}\boldsymbol{k},$$

which satisfies $P^r_{\alpha\beta} = -P^r_{\beta\alpha}$.

Proof. Observe that

$$\frac{i\omega_{mn}(\boldsymbol{k})}{\omega + \omega_{mn}(\boldsymbol{k})} = i + \frac{-i\omega}{\omega + \omega_{mn}(\boldsymbol{k})}, \quad \frac{-i\omega_{mn}(\boldsymbol{k})}{\omega - \omega_{mn}(\boldsymbol{k})} = i + \frac{-i\omega}{\omega - \omega_{mn}(\boldsymbol{k})}.$$

Then it is easy to see that Proposition 6.2 along with (6.3) implies $\widehat{\boldsymbol{g}}(\omega) = (-i\omega)\widehat{\boldsymbol{f}}(\omega)$, and Proposition 6.2 along with (6.4) implies $\widehat{N}_{\alpha\beta}(\omega) - \langle \rho_0 \rangle_{\boldsymbol{z}} \delta_{\alpha\beta} = (-i\omega)\widehat{M}_{\alpha\beta}(\omega)$.

Moreover, Proposition 6.2 also implies

$$\begin{aligned}
\widehat{M}_{\alpha\beta}(\omega) =& - \widehat{R}_{\alpha\beta}(\omega), \\
\widehat{R}_{\alpha\beta}(\omega) =& (-i\omega)\widehat{P}_{\alpha\beta}(\omega) \\
& + i \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \overline{\langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{k}_\alpha} | u_{m,\boldsymbol{k}}\rangle} \langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{k}_\beta} | u_{m,\boldsymbol{k}}\rangle \,\mathrm{d}\boldsymbol{k} \\
& - i \sum_{n\le Z}\sum_{m>Z} \fint_{\Gamma^*} \langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{k}_\alpha} | u_{m,\boldsymbol{k}}\rangle \overline{\langle u_{n,\boldsymbol{k}} | i\partial_{\boldsymbol{k}_\beta} | u_{m,\boldsymbol{k}}\rangle} \,\mathrm{d}\boldsymbol{k} \\
=& (-i\omega)\big(\widehat{P}_{\alpha\beta}(\omega) - P^r_{\alpha\beta}\big).
\end{aligned}$$

□

Note that

$$\partial_{\boldsymbol{x}_\alpha}\partial_{\boldsymbol{x}_\beta}\widehat{U_0} = \partial_{\boldsymbol{x}_\beta}\partial_{\boldsymbol{x}_\alpha}\widehat{U_0}, \qquad P^r_{\alpha\beta} = -P^r_{\beta\alpha},$$

one knows that the equation (6.1) will remain the same if we redefine

$$A_{\alpha\beta} = \widehat{P}_{\alpha\beta} - P^r_{\alpha\beta}(\omega) - \langle \widehat{\boldsymbol{f}}^*_\alpha \mathcal{V}(I - \widehat{\chi}_\omega \mathcal{V})^{-1} \widehat{\boldsymbol{f}}_\beta \rangle_{\boldsymbol{z}}.$$

Then Lemma 6.4 implies

$$B_{\alpha\beta} = (-i\omega)A_{\alpha\beta}, \quad D_{\alpha\beta} = (-i\omega)C_{\alpha\beta}, \quad C_{\alpha\beta} = -B_{\alpha\beta}.$$

By defining $\widehat{\boldsymbol{E}} = -\nabla_{\boldsymbol{x}}\widehat{U_0} + i\omega\widehat{\boldsymbol{A}_0}$, $\widehat{\boldsymbol{B}} = \nabla_{\boldsymbol{x}} \times \widehat{\boldsymbol{A}_0}$, the equations (6.1)-(6.2) along with $\nabla_{\boldsymbol{x}} \cdot \widehat{\boldsymbol{A}_0} = 0$ produce (3.29)-(3.32). This completes the derivation of the main result in Section 3.3.

7. Conclusion

One unsatisfactory aspect of this work is that it is limited to short time scales. In fact, the behavior at longer time scales is still very much of a mystery, even from the viewpoint of formal asymptotics. This is very unsettling. The main technical difficulty is the lack of local charge neutrality and the huge potential generated as a result.

There are other important issues that remain. These include the inclusion of spin, the interaction with lattice dynamics, defects, instabilities, etc.

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