

On the equivalence of the semiclassical theory and the response theory

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It is commonly believed that the response theory can give quantum correction (or interband coherent effects) to the semiclassical theory, while both formulations essentially are perturbatively solving the time-dependent Schrödinger equation in a periodic potential probed by the electric field within the independent-particle approximation. Herein, by extending the semiclassical theory under an AC uniform electric field to the nonlinear regime, we show that up to the second order of the electric field, the AC semiclassical theory is equivalent to the response theory in the absence of relaxation. Remarkably, this equivalence can be inherited when the relaxation is incorporated into the response theory, particularly by taking the semiclassical results with a finite relaxation time obtained by solving the Boltzmann equation as a benchmark. Specifically, in the linear order of the electric field, we show that both formulations deliver the Drude current, the intrinsic anomalous Hall current that is driven by the Berry curvature, and the intrinsic displacement current that is driven by the quantum metric, where the last one can only appear in AC transport. Furthermore, in the second order of the electric field, we show that the familiar second-order transport effects, including the nonlinear Drude current, the extrinsic nonlinear Hall current that is driven by the Berry curvature dipole, and the intrinsic nonlinear Hall current that is driven by the quantum metric dipole, can be derived by both formulations. In addition, we show that the intrinsic nonlinear Ohmic current that is also driven by the quantum metric dipole, as first formulated by the response theory, can be derived from the semiclassical theory by taking the second-order semiclassical wavepacket energy into account. Moreover, we show that both formulations can give the extrinsic and intrinsic nonlinear displacement currents, both of which can only appear in AC transport and are not reported yet. Notably, we find that the extrinsic nonlinear displacement current is also solely driven by the quantum metric dipole but scales with a dimensionless constant $\omega\tau$, where ω and τ stand for the driving frequency and the relaxation time, respectively. As a result, both the quantum-metric-dipole driven intrinsic nonlinear Hall and Ohmic currents under an AC electric field can be significantly enhanced by this extrinsic nonlinear displacement current when $\omega\tau \gg 1$. Besides, we note that the $\mathcal{T}$ -even ($\mathcal{T}$, time reversal) intrinsic nonlinear displacement current can coexist with the Berry-curvature-dipole driven extrinsic nonlinear Hall effect under an AC electric field. As a consequence of this equivalence, (i) we conclude that the semiclassical theory does not ignore interband coherent effects compared to the response theory; (ii) we show that the intrinsic nonlinear Ohmic current and the nonreciprocal magnetoresistance represent the same physics, both of which arise from the second-order semiclassical wavepacket energy correction; (iii) we suggest that the energy used in the equilibrium Fermi distribution in the semiclassical theory is the unperturbed band energy by assuming that both formulations give the same intrinsic responses; (iv) we argue that the scheme of introducing relaxation in the response theory by modifying the Liouville equation for the nonequilibrium density matrix needs to be reconsidered. Our work clarifies the conceptual puzzles encountered in the emergent field of *nonlinear Hall effects* and highlights that the displacement nonlinear current responses unique to the AC transport can be important especially when a high-frequency AC driving field is applied.

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I. INTRODUCTION

Understanding the response or transport behaviors of Bloch electrons in crystalline solids under the electromagnetic field is one of the oldest but still most active themes in condensed matter physics [1], which may be traced back to the phenomenological Drude model for metal with a finite Fermi surface. Along this way, by treating the Bloch electron as a wavepacket which satisfies the time-dependent Schrödinger equation, the semi-classical equations of motion under the static electric field [2–5] ($e = \hbar = 1$)

$$\dot{\mathbf{r}} = -\dot{\mathbf{k}} \times \boldsymbol{\Omega} + \partial_{\mathbf{k}}\epsilon, \quad (1)$$

$$\dot{\mathbf{k}} = -\mathbf{E}, \quad (2)$$

are proposed to evaluate the velocity for the position center $\mathbf{r}$ of the wavepacket in phase space. Furthermore, by combining with the Boltzmann equation for the nonequilibrium distribution function $\bar{f}_n$ for the n th band,

$$\mathbf{E} \cdot \nabla_{\mathbf{k}} \bar{f}_n - \partial_t \bar{f}_n = (\bar{f}_n - f_n)/\tau, \quad (3)$$

the current at the linear and nonlinear regimes can be calculated [6–14], where the scattering is usually introduced by adopting the relaxation time (τ) approximation. Importantly, the linear and nonlinear conductivity tensors usually are expressed by the quantum Bloch wavefunction and thereby the detected current can manifest the quantum geometry [15–20] and topological properties [21–23] of quantum materials, as can be achieved by steering the symmetry that bridges the quantum geometric quantities and the conductivity tensors.

In particular, Eqs.(1-3), especially when combining with the well-developed first-principles material calculations [24], offers one of the most standard and successful theoretical tools to quantitatively appreciate the exotic linear and nonlinear transport experiments, such as the intrinsic anomalous Hall effect [25–28] in ferromagnetic metals and the extrinsic nonlinear Hall effect in nonmagnetic quantum materials [29–31], where the Berry curvature $\boldsymbol{\Omega}$ from Eq.(1) plays a pivotal role [4]. Recently, based on the extended semiclassical theory [7], one further realizes that the counterpart of the Berry curvature in the quantum geometric tensor—quantum metric, can also play a critical role, such as in the intrinsic second-order nonlinear Hall effect [32–38] and the extrinsic third-order nonlinear Hall effect [9, 10]. These progresses spark the quantum geometric engineering in topological quantum materials [15], where the traditional paradigm that the response of a solid to a weak electromagnetic field is fully determined by the band dispersion is shifting.

Compared to the transport in metal, the response properties of Bloch electrons in insulators with a vanishing Fermi surface are very often explored by irradiating light (an AC electric field), which may be traced back to the phenomenological Lorentz model [39] for the dielectric solids. Up to date, the more standard and mature approach used to tackle with the same problem is the response theory [40–51], where the valence electrons by absorbing a photon to form a *resonant* conducting current can be well captured. Within the framework of the response theory, the linear and nonlinear currents are obtained by solving the (quantum) Liouville equation for the nonequilibrium density matrix ρ [43–45],

$$i\partial_t \rho = [H, \rho], \quad (4)$$

where $H = H_0 + H_1$ with H_0 being the solved periodic Hamiltonian and $H_1 = \mathbf{E}(t) \cdot \mathbf{r}$ the perturbed electric field at the length gauge [43, 44, 52].

Similarly, also by combining with the well-developed first-principles material calculations, the expressions derived by the response theory can also be used to quantitatively evaluate the current in realistic materials [47–50], such as the shift and injection current responses [43, 44]. Importantly, a number of transport experiments under light illumination can be appreciated by those first-principles calculations based on the response theory [53–58]. In addition, these exotic responses can also be employed to diagnose the quantum geometric properties of Bloch electrons in quantum materials [15–20].

Despite their distinct starting points, the semiclassical theory and the response theory has been overlapped in many aspects [59–68]. For instance, both formulations give the same conductivity for the linear responses [59–62]. In addition, all the nonlinear (charge) response relations derived by the semiclassical theory, which contains the extrinsic nonlinear Drude current [7], the extrinsic nonlinear Hall current [6], and the intrinsic nonlinear Hall current [7], have been reproduced by the response theory under the DC limit [64].

Recently, two intrinsic second-order nonlinear responses, which are suggested to be beyond the semiclassical results given in the pioneering paper Ref. [7], are proposed. The first one is the intrinsic second-order nonlinear Ohmic current, which was predicted in Refs. [65, 66] within the response theory and was attributed to the interband coherent correction compared to the semiclassical results in Ref. [7]. The second one is the non-reciprocal magnetoresistance (intrinsic second-order nonlinear longitudinal current), which was predicted by the Kohn-Luttinger method [69] and can be unified with the intrinsic nonlinear Hall current proposed in Ref. [7] from the quantum geometry. Nevertheless, we wish to mention that Ref. [7] has suppressed the intrinsic transport due to the field-induced group velocity, which arises from the second-order wavepacket energy correction [70, 71] and can contribute to both the longitudinal and transverse intrinsic nonlinear currents. This naturally poses an intriguing question: is it possible that the second-order wavepacket energy is responsible for the missed nonlinear physics discussed above? And both formulations in fact are equivalent even at the second order?

In addition to the overlap, the semiclassical theory and the response theory in their own regard also meet some challenges. For the semiclassical theory, as discussed above, the external electric field can induce a second-order energy correction and thereby can deliver a field-induced group velocity in the semiclassical equations of motion. However, it is not clear that whether or not such an energy correction should be placed into the Fermi distribution function. Specifically, in terms of the semiclassical theory, the intrinsic current under the static electric field is given by [8]

$$\mathbf{J} = \sum_n \int_k f_n [\bar{\mathbf{v}}_n + \mathbf{E} \times \bar{\boldsymbol{\Omega}}_n], \quad (5)$$

where $\bar{\mathbf{v}}_n = \nabla_k \bar{\epsilon}_n$ with $\bar{\epsilon}_n = \epsilon_n + \epsilon_n^{(2)}$ (ϵ_n , the unperturbed energy; $\epsilon_n^{(2)}$, the second-order energy correction [70, 71]) is the group velocity and $\bar{\boldsymbol{\Omega}}_n$ is the Berry curvature accurate up to the first order of the electric field. Since the energy is the argument of the equilibrium Fermi distribution function, it becomes conceptually involved that which one, namely $f_n(\epsilon_n)$ or $f_n(\bar{\epsilon}_n)$ [8, 9], should be used in Eq.(5) especially when evaluating the nonlinear responses.

Furthermore, we note that the semiclassical equations of motion Eqs. (1-2) under the static electric field have

been directly extended to calculate the time-dependent high-frequency responses [6, 77–79]. Meanwhile, almost all the experiments [29–31, 34–36] to verify these semiclassical proposals are performed by applying an AC driving field. However, the complete semiclassical equations of motion under the AC (time-dependent) electric field read [2–4]

$$\dot{\mathbf{r}} = -\dot{\mathbf{k}} \times \boldsymbol{\Omega} + \partial_{\mathbf{k}} \epsilon - \boldsymbol{\Omega}_{\mathbf{k}t}, \quad (6)$$

$$\dot{\mathbf{k}} = -\mathbf{E}(t), \quad (7)$$

where the mixed Berry curvature $\boldsymbol{\Omega}_{\mathbf{k}t}$ defined in momentum-time space, like the conventional Berry curvature $\boldsymbol{\Omega}$, also shows a critical role, such as in the adiabatic charge pumping [80], the emergent electromagnetic induction [81], and the linear intrinsic displacement current [62]. It has been well known that the electric field can lead to a correction to the conventional Berry curvature and further induces the intrinsic nonlinear Hall effect [7, 32–35], but how the mixed Berry curvature can be modified by the electric field and whether or not this modification can bring up any unexplored physics remain elusive. Besides, an explicit comparison between the AC semiclassical theory and the response theory with a finite frequency has not been conducted, particularly at the second order of the electric field.

On the other hand, for the response theory, the inclusion of relaxation usually was achieved by phenomenologically modifying the Liouville equation as [72, 73]

$$i\partial_t \rho = [H, \rho] - \frac{i}{\tau}(\rho - \rho^{(0)}), \quad (8)$$

where $\rho^{(0)}$ is the equilibrium density matrix. Although Eq. (8) has been used in many studies to discuss the nonlinear responses [74–76], the additional term in Eq. (8) has not been fully justified, as far as we know. In particular, if Eq. (8) does not work, how to introduce the relaxation in the response theory is unknown.

To address these questions, we extend the semiclassical theory under an AC uniform electric field up to the second order of the electric field. We show that up to the second order of electric field, the AC semiclassical theory is equivalent to the response theory in the clean limit. Remarkably, this equivalence can also be expected when the relaxation, which is introduced by solving the Boltzmann equation in the semiclassical results and is responsible for regulating the divergent DC conductivity in the clean limit, is incorporated into the response theory by regulating the divergent DC conductivity on the same footing. In particular, in the linear order of the electric field, we show that both formulations can give the linear Drude current, intrinsic anomalous Hall current, and the displacement current, where the last one arising from the time variation of the first-order *positional shift* [7] can only be induced by an AC electric field. Furthermore, in the second order of the electric field, we show that the familiar second-order transport effects, including the nonlinear Drude current, the extrinsic and intrinsic nonlinear Hall currents, the intrinsic nonlinear Ohmic current,

can be derived with both formulations. In addition, we find that the extrinsic and intrinsic nonlinear displacement currents which arises from the time variation of the first-order and the second-order *positional shifts* [7, 82], respectively, can also be derived with both formulations. Remarkably, we find that the extrinsic nonlinear displacement current features the same quantum geometric origin with the intrinsic nonlinear Hall and Ohmic currents but scales with a dimensionless constant $\omega\tau$, where ω stands for the driving frequency. As a result, both the intrinsic nonlinear Hall and Ohmic currents can be largely amplified by the extrinsic nonlinear displacement current, particularly under an AC driving field with $\omega\tau \gg 1$. Besides, we note that the intrinsic nonlinear displacement conductivity features the $\mathcal{T}$ -even ($\mathcal{T}$, time reversal) property so that this nonlinear current can coexist with the $\mathcal{T}$ -even Berry-curvature-dipole driven extrinsic nonlinear Hall effect under an AC electric field.

On top of the equivalence between both formulations, (i) we conclude that the semiclassical theory does not ignore any interband coherent effects compared to the response theory; (ii) we find that the intrinsic nonlinear Ohmic current and the nonreciprocal magnetoresistance (that is added into the second-order semiclassical results with the Kohn-Luttinger method) represent the same physics, both of which arise from the second-order wavepacket energy correction of the semiclassical theory; (iii) we suggest that the energy used in the equilibrium Fermi distribution in the semiclassical theory is the unperturbed band energy since both formulations are assumed to give the same intrinsic responses; (iv) we argue that the scheme of introducing relaxation in the response theory by modifying the Liouville equation for the density matrix needs to be reconsidered since Eq.(8) can give a linear extrinsic Hall current in $\mathcal{T}$ -invariant systems, as forbidden by the Onsager reciprocal relation [12, 28, 83].

The remainder of this paper is organized as follows. In section II, we develop the second-order AC semiclassical theory with the wavepacket method. In section III, we derive the current density up to the second order of the electric field with the extended AC semiclassical theory and the response theory, respectively. In section IV, we explicitly show the equivalence of the current density derived by the AC semiclassical theory and the response theory up to the second order of the electric field, for both the zero-frequency and finite-frequency scenarios. Based on this equivalence, in section V we clarify the inconsistent overlap between both formulations discussed above and address the challenges confronted in each theory. Furthermore, in section VI, we discuss the nonlinear displacement currents unique to the AC transport, which are derived by both formulations and contain both the intrinsic and extrinsic contributions. In section VII, we summarize the main results and briefly outline the future direction. Finally, an Appendix VIII is given, which includes the calculation details for the conductivity tensors with the response theory, the solution for the Boltzmann equation, some calculation details for devel-

oping the second-order AC semiclassical theory and for showing the equivalence between both formulations, and the calculation details for the second-order energy with the Kohn-Luttinger method.

II. AC SEMICLASSICAL THEORY

In this section, we extend the AC semiclassical theory under a time-dependent and uniform electric field to the second order of the driving field. In detail, we construct the wavepacket accurate up to the second order of the AC electric field and then evaluate the position center for the wavepacket, where the key expressions for the *positional shift* up to the second order of the AC electric field are obtained. In addition, we derive the equations of motions for the wavepacket centers (including momentum and position) find that the electric field can also induce a correction to the mixed Berry curvature that is solely decided by the time variation of the second-order *positional shift*. Finally, we estimate the wavepacket energy accurate up to the second order of the AC electric field and we find that this energy correction can also be expressed by the first-order *positional shift*, like the extended DC semiclassical theory.

A. The wavepacket accurate up to the second order

Following the spirit of the semiclassical wavepacket theory [4, 7], we focus on the n th band to construct the wavepacket as follows ($e = \hbar = 1$):

$$|W\rangle = \int_{\mathbf{p}} e^{i\mathbf{p}\cdot\mathbf{r}} \left(C_n(\mathbf{p})|u_n\rangle + \sum_{l \neq n} \bar{C}_l(\mathbf{p})|u_l\rangle \right), \quad (9)$$

where $\int_{\mathbf{p}} \equiv \int d\mathbf{p}/(2\pi)^d$ with d the system dimension, $|u_n\rangle \equiv |u_n(\mathbf{p})\rangle$ is periodic part of the Bloch state, $C_n(\mathbf{p})$ is the zero-order amplitude and $\bar{C}_l(\mathbf{p})$ stands for the correction (indicated with a bar above C_l) induced by the external electric field (Throughout this work, we will only consider the electric field). Here we assume that [4]

$$|C_n(\mathbf{p})|^2 = \delta(\mathbf{p} - \mathbf{p}_c) \quad (10)$$

with $\mathbf{p}_c$ the momentum center to normalize the wavepacket up to the first order [4]. Note that Eq.(10) is useful in evaluating various quantities related to the wavepacket.

As the superposition of Bloch states, the wavepacket obeys the time-dependent Schrödinger equation

$$i\partial_t|W\rangle = H|W\rangle, \quad (11)$$

where $\partial_t = \partial/\partial t$, $H = H_0 + H_1$ with H_0 the periodic crystal Hamiltonian, which gives $H_0|u_n\rangle = \epsilon_n|u_n\rangle$,

and $H_1 = \mathbf{E}(t) \cdot \mathbf{r}$ the perturbed Hamiltonian due to the applied AC electric field $\mathbf{E}(t)$. Particularly, $\mathbf{E}(t) = \sum_{\omega_1} \mathbf{E} e^{-i(\omega_1 + i\xi)t}$ with $\omega_1 = \pm\omega$, which is adiabatically

turned on due to $\xi \rightarrow 0^+$. By multiplying $\langle u_m(\mathbf{p}) | e^{-i\mathbf{p} \cdot \mathbf{r}}$ on both sides of Eq.(11), we find

$$\begin{aligned} \int_{\mathbf{p}'} e^{i(\mathbf{p}' - \mathbf{p}) \cdot \mathbf{r}} \left(i\dot{C}_n(\mathbf{p}') \langle u_m | u'_n \rangle + \sum_{l \neq n} i\dot{\bar{C}}_l(\mathbf{p}') \langle u_m | u'_l \rangle \right) &= \int_{\mathbf{p}'} e^{i(\mathbf{p}' - \mathbf{p}) \cdot \mathbf{r}} \left(C_n(\mathbf{p}') \langle u_m | H_0 | u'_n \rangle + \sum_{l \neq n} \bar{C}_l(\mathbf{p}') \langle u_m | H_0 | u'_l \rangle \right) \\ &+ \int_{\mathbf{p}'} e^{i(\mathbf{p}' - \mathbf{p}) \cdot \mathbf{r}} \left(C_n(\mathbf{p}') \langle u_m | H_1 | u'_n \rangle + \sum_{l \neq n} \bar{C}_l(\mathbf{p}') \langle u_m | H_1 | u'_l \rangle \right), \end{aligned} \quad (12)$$

where $\dot{C}_n = \partial_t C_n$ and $|u'_n\rangle = |u_n(\mathbf{p}')\rangle$. At this stage, using the orthogonal relation between Bloch states

$$\langle u_m | e^{i(\mathbf{p}' - \mathbf{p}) \cdot \mathbf{r}} | u'_n \rangle = \delta(\mathbf{p}' - \mathbf{p}) \delta_{mn} \quad (13)$$

and the Bloch representation of position operator $\mathbf{r}$ [43, 44]

$$\langle u_m | e^{i(\mathbf{p}' - \mathbf{p}) \cdot \mathbf{r}} \mathbf{r} | u'_n \rangle = [\delta_{mn} i\partial_{\mathbf{p}} + \mathcal{A}_{mn}] \delta(\mathbf{p} - \mathbf{p}'), \quad (14)$$

where $\partial_{\mathbf{p}} \equiv \partial/\partial\mathbf{p}$ and $\mathcal{A}_{nm} \equiv \langle u_n | i\partial_{\mathbf{p}} | u_m \rangle$, when $m = n$ Eq.(12) becomes [84]

$$i\dot{C}_n = \epsilon_n C_n + \mathbf{E} \cdot (i\partial_{\mathbf{p}} + \mathcal{A}_n) C_n + \sum_l \bar{C}_l \mathbf{E} \cdot \mathbf{r}_{nl}, \quad (15)$$

where $\mathcal{A}_n \equiv \mathcal{A}_{nn}$ is the intraband Berry connection and $\mathbf{r}_{nl} \equiv \mathcal{A}_{nl}$ for $n \neq l$ the interband Berry connection. However, when $m \neq n$ Eq.(12) reduces into

$$\begin{aligned} i\dot{\bar{C}}_m &= \epsilon_m \bar{C}_m + \mathbf{E} \cdot \mathbf{r}_{mn} C_n + \mathbf{E} \cdot (i\partial_{\mathbf{p}} + \mathcal{A}_m) \bar{C}_m \\ &+ \sum_l \bar{C}_l \mathbf{E} \cdot \mathbf{r}_{ml}, \end{aligned} \quad (16)$$

Eq.(16), together with Eq.(15), fully determines the unknown coefficient $\bar{C}_l$ in the wavepacket Eq.(9). Particularly, let $\bar{C}_m = M_{mn} C_n$, we find that Eq.(16) becomes

$$\begin{aligned} i\dot{M}_{mn} C_n + M_{mn} i\dot{C}_n &= \epsilon_m M_{mn} C_n + \mathbf{E} \cdot \mathbf{r}_{mn} C_n \\ &+ \mathbf{E} \cdot (i\partial_{\mathbf{p}} + \mathcal{A}_m) M_{mn} C_n + \sum_l \mathbf{E} \cdot \mathbf{r}_{ml} M_{ln} C_n. \end{aligned} \quad (17)$$

Then by substituting Eq.(15) into this equation and eliminating C_n , we finally obtain

$$\begin{aligned} \dot{M}_{mn} + i\epsilon_{mn} M_{mn} &= -i\mathbf{E} \cdot \mathbf{r}_{mn} + \mathbf{E} \cdot \mathcal{D}_{mn}^p M_{mn} \\ &- i \sum_l \mathbf{E} \cdot (\mathbf{r}_{ml} - M_{mn} \mathbf{r}_{nl}) M_{ln}, \end{aligned} \quad (18)$$

where $\mathcal{D}_{mn}^p \equiv \partial_{\mathbf{p}} - i(\mathcal{A}_m - \mathcal{A}_n)$ is the $U(1)$ gauge-invariant derivative and $\epsilon_{mn} = \epsilon_m - \epsilon_n$ the energy difference. Eq.(18) can be solved iteratively by writing

$M_{mn} = \sum_i M_{mn}^{(i)}$ with $i \geq 1$ and $M_{mn}^{(i)} \propto E_{\alpha}^{(i)}$. For $i = 1$, we find

$$\dot{M}_{mn}^{(1)} + i\epsilon_{mn} M_{mn}^{(1)} = -iE_{\alpha}^{\omega_1} r_{mn}^{\alpha}, \quad (19)$$

where $E_{\alpha}^{\omega_1} = E_{\alpha} e^{-i(\omega_1 + i\xi)t}$ with $\omega_1 = \pm\omega$ (Throughout this work, the summation over frequency is suppressed unless otherwise stated) and the Einstein summation convention for the repeated Greek alphabets has been assumed, and thereby

$$M_{mn}^{(1)} = \frac{r_{mn}^{\alpha} E_{\alpha}^{\omega_1}}{\omega_1 - \epsilon_{mn} + i\xi}. \quad (20)$$

Similarly, for $i = 2$, we find:

$$\begin{aligned} \dot{M}_{mn}^{(2)} + i\epsilon_{mn} M_{mn}^{(2)} &= E_{\beta} \mathcal{D}_{mn}^{\beta} M_{mn}^{(1)} - i \sum_l E_{\beta}^{\omega_2} r_{ml}^{\beta} M_{ln}^{(1)}, \end{aligned} \quad (21)$$

where $\mathcal{D}_{mn}^{\beta} = \partial_{\beta} - i(\mathcal{A}_m^{\beta} - \mathcal{A}_n^{\beta})$ with $\partial_{\beta} = \partial/\partial p_{\beta}$. By solving Eq.(21) we obtain

$$\begin{aligned} M_{mn}^{(2)} &= i\mathcal{D}_{mn}^{\beta} \left(\frac{r_{mn}^{\alpha}}{\omega_1 - \epsilon_{mn} + i\xi} \right) \frac{E_{\alpha}^{\omega_1} E_{\beta}^{\omega_2}}{\omega_{\Sigma} - \epsilon_{mn} + 2i\xi} \\ &+ \sum_l \frac{r_{ml}^{\beta} r_{ln}^{\alpha} E_{\alpha}^{\omega_1} E_{\beta}^{\omega_2}}{(\omega_1 - \epsilon_{ln} + i\xi)(\omega_{\Sigma} - \epsilon_{mn} + 2i\xi)}, \end{aligned} \quad (22)$$

where $\omega_{\Sigma} = \omega_1 + \omega_2$.

With $M_{mn}^{(1)}$ and $M_{mn}^{(2)}$, the wavepacket accurate up to the second order of the electric field is given by

$$\begin{aligned} |W\rangle &= \int_{\mathbf{p}} e^{i\mathbf{p} \cdot \mathbf{r}} C_n(\mathbf{p}) \left[|u_n\rangle + \sum_{m \neq n} \left(M_{mn}^{(1)} + M_{mn}^{(2)} \right) |u_m\rangle \right] \\ &\equiv |W_0\rangle + |W_1\rangle + |W_2\rangle, \end{aligned} \quad (23)$$

where $\bar{C}_l = \sum_i C_m^{(i)}$ with $C_m^{(i)} = M_{mn}^{(i)} C_n$ has been used and $|W_i\rangle \propto E_{\alpha}^{(i)}$.

Note that the wavepacket will be used to evaluate the Lagrangian so we need to normalize it up to the second

order of the electric field, which in fact has been carefully considered in the previous nonlinear semiclassical studies [71, 85]. Particularly, we have:

$$\begin{aligned} |\bar{W}\rangle &= \frac{|W\rangle}{\sqrt{\langle W|W\rangle}} = \frac{|W\rangle}{\sqrt{1 + \langle W_1|W_1\rangle + \mathcal{O}(E_\alpha^3)}} \\ &= [1 + \delta + \mathcal{O}(E_\alpha^3)] |W\rangle, \end{aligned} \quad (24)$$

where we have performed a Taylor expansion and obtained [85]

$$\delta \equiv -\frac{1}{2}\langle W_1|W_1\rangle = -\frac{1}{2}\sum_m M_{mn}^{(1)}(\mathbf{p}_c)M_{mn}^{(1)*}(\mathbf{p}_c), \quad (25)$$

by using Eq.(10) and Eq.(23). Finally, the normalized AC wavepacket accurate up to the second order of the electric field can be expressed as

$$|\bar{W}\rangle = (1 + \delta)|W_0\rangle + |W_1\rangle + |W_2\rangle, \quad (26)$$

which constitutes the starting point to formulate the AC semiclassical theory particularly accurate up to the second order of the electric field.

B. The position center for the wavepacket

With the constructed wavepacket $|\bar{W}\rangle$, the position center $\mathbf{r}_c$ defined by $\mathbf{r}_c \equiv \langle \bar{W}|\mathbf{r}|\bar{W}\rangle$ is found to be (see Appendix A 1)

$$\mathbf{r}_c = \partial_{\mathbf{p}_c}\gamma + \mathcal{A}_n + \mathbf{a}_n^{(1)} + \mathbf{a}_n^{(2)}, \quad (27)$$

where $\gamma = -\text{Arg}(C_n)$ and we have suppressed the dependence on $\mathbf{p}_c$ for all quantities for brevity. In addition, here $\mathbf{a}_n^{(i)}$ is the i th *positional shift* given by

$$\mathbf{a}_n^{(1;\lambda)} = 2\text{Re}\sum_m \frac{r_{nm}^\lambda r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} E_\alpha^{\omega_1}, \quad (28)$$

and

$$\begin{aligned} \mathbf{a}_n^{(2;\lambda)} &= \text{Re}\sum_{ml} \frac{2r_{nm}^\lambda r_{ml}^\beta r_{ln}^\alpha}{(\omega_1 - \epsilon_{ln} + i\xi)(\omega_\Sigma - \epsilon_{mn} + 2i\xi)} E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ &+ \sum_{ml} \frac{-r_{nl}^\beta r_{lm}^\lambda r_{mn}^\alpha}{(\omega_2 - \epsilon_{nl} + i\xi)(\omega_1 - \epsilon_{mn} + i\xi)} E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ &+ \text{Re}\sum_m i\mathcal{D}_{mn}^\beta \left(\frac{r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} \right) \frac{2r_{nm}^\lambda E_\alpha^{\omega_1} E_\beta^{\omega_2}}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} \\ &+ \text{Re}\sum_m i\mathcal{D}_{nm}^\lambda \left(\frac{r_{nm}^\alpha}{\omega_1 - \epsilon_{nm} + i\xi} \right) \frac{r_{mn}^\beta E_\alpha^{\omega_1} E_\beta^{\omega_2}}{\omega_2 - \epsilon_{mn} + i\xi}, \end{aligned} \quad (29)$$

where again the Einstein summation convention for the repeated Greek alphabets is assumed and the summation over frequency has been suppressed. Note that under $U(1)$ gauge transformation $|u_n\rangle \rightarrow e^{i\phi_n}|u_n\rangle$, $\mathcal{A}_n^\alpha \rightarrow \mathcal{A}_n^\alpha - \partial_\alpha\phi_n$, $r_{nm}^\alpha \rightarrow e^{i(\phi_m - \phi_n)}r_{nm}^\alpha$, it is easy to check that both $\mathbf{a}_n^{(1;\lambda)}$ and $\mathbf{a}_n^{(2;\lambda)}$ are gauge invariant, as they must be.

C. The equations of motion for the wavepacket

With the normalized AC wavepacket $|\bar{W}\rangle$, the equations of motion for the wavepacket centers $\mathbf{p}_c$ and $\mathbf{r}_c$ can also be derived. To that purpose, we first evaluate the Lagrangian for the wavepacket, which is defined as:

$$\mathcal{L} \equiv \langle \bar{W} | (i\partial_t - H) | \bar{W} \rangle = \langle W_0 | i\partial_t | W_0 \rangle - \bar{\epsilon}_n, \quad (30)$$

where $\bar{\epsilon}_n \equiv \langle W_0 | i\partial_t | W_0 \rangle - \langle \bar{W} | (i\partial_t - H) | \bar{W} \rangle$ is the wavepacket energy (accurate up to the second order of the electric field for our purpose) that will be calculated in the subsequent subsection. Note that

$$\langle W_0 | i\partial_t | W_0 \rangle = \partial_t \gamma(\mathbf{p}_c), \quad (31)$$

and therefore

$$\begin{aligned} \mathcal{L} &= \partial_t \gamma - \bar{\epsilon}_n = \frac{d}{dt}\gamma - \partial_{\mathbf{p}_c}\gamma \cdot \dot{\mathbf{p}}_c - \bar{\epsilon}_n \\ &= -\dot{\mathbf{k}}_c \cdot (\mathbf{r}_c - \bar{\mathcal{A}}_n) - \bar{\epsilon}_n, \end{aligned} \quad (32)$$

where we have used Eq.(27) and defined $\bar{\mathcal{A}}_n \equiv \mathcal{A}_n + \mathbf{a}_n^{(1)} + \mathbf{a}_n^{(2)}$. Here we have dropped the unimportant total time derivative $d\gamma/dt$ in the Lagrangian and replaced $\mathbf{p}_c$ with $\mathbf{k}_c$ in the final result. In addition, we have assumed that the eigenstates of Hamiltonian do not explicitly depend on time so that $\mathcal{A}_n^t \equiv \langle u_n | \partial_t | u_n \rangle = 0$ [4]. Next, using the Euler-Lagrange equations,

$$\frac{d}{dt} \frac{\partial \mathcal{L}}{\partial \dot{\mathbf{k}}_c} = \frac{\partial \mathcal{L}}{\partial \mathbf{k}_c}, \quad \frac{d}{dt} \frac{\partial \mathcal{L}}{\partial \dot{\mathbf{r}}_c} = \frac{\partial \mathcal{L}}{\partial \mathbf{r}_c}, \quad (33)$$

we find that

$$\dot{\mathbf{r}} = -\dot{\mathbf{k}} \times \bar{\boldsymbol{\Omega}}_n + \bar{\mathbf{v}}_n + \partial_t \bar{\mathcal{A}}_n, \quad (34)$$

$$\dot{\mathbf{k}} = -\mathbf{E}(t), \quad (35)$$

where we have further suppressed the subscript c on $\mathbf{r}_c$ and $\mathbf{k}_c$. In addition, we have used $\partial \bar{\epsilon}_n / \partial \mathbf{r}_c = \mathbf{E}(t)$ from Eq. (36) and defined $\partial \bar{\epsilon}_n / \partial \mathbf{k}_c \equiv \bar{\mathbf{v}}_n$ and $\bar{\boldsymbol{\Omega}}_n \equiv \partial / \partial \mathbf{k}_c \times \bar{\mathcal{A}}_n$. Note that Eqs. (34-35) are formally the same as the first-order or linear AC semiclassical theory developed by G. Sundaram and Q. Niu in Ref. [3].

We wish to remark that the equations of motion for the AC semiclassical theory is the same as the DC semiclassical theory except for the last term in Eq.(34). In fact, this term is only contributed by the time variation of *positional shift* since $\mathcal{A}_n$ is time-independent, especially, $\partial_t \bar{\mathcal{A}}_n = \partial_t(\mathbf{a}_n^{(1)} + \mathbf{a}_n^{(2)})$. Remarkably, this additional term will give the displacement current under the time-dependent external electric field [62] and is usually overlooked even in the AC transport. Interestingly, we note that the *positional shift* also enters the first term of Eq.(34), which gives the anomalous velocity $\mathbf{E} \times \boldsymbol{\Omega}_n^{(i)}$ at the $(i+1)$ th order of the electric field since $\boldsymbol{\Omega}_n^{(i)} = \partial_{\mathbf{k}} \times \mathbf{a}_n^{(i)}$ with $i \geq 1$. As a result, the first-order *positional shift* shows up only in the second-order DC semiclassical theory, whereas the same *positional shift* must

be calculated in the first-order AC semiclassical theory. In addition, the energy correction for the wavepacket particularly at second order of the electric field in fact is determined by the first-order *positional shift*, as will be shown below.

D. The wavepacket energy

To complete the AC semiclassical theory particularly accurate up to the second order of the electric field, we now derive the wavepacket energy $\bar{\epsilon}_n$ accurate up to the same order, which determines the group velocity appeared in Eq.(34). In particular, we find (see Appendix A 2),

$$\begin{aligned}\bar{\epsilon}_n &\equiv \langle W_0 | i\partial_t | W_0 \rangle - \langle \bar{W} | (i\partial_t - H) | \bar{W} \rangle \\ &= \epsilon_n + \mathbf{E} \cdot \mathbf{r}_c + \epsilon_n^{(2)} + \mathcal{O}(E_\alpha^3)\end{aligned}\quad (36)$$

where

$$\begin{aligned}\epsilon_n^{(2)} &\equiv \text{Re} \sum_m \left[\left(i\partial_t M_{mn}^{(1)} \right) M_{mn}^{(1)*} + \epsilon_{mn} M_{mn}^{(1)} M_{mn}^{(1)*} \right] \\ &= -\text{Re} \sum_m \frac{(\omega_1 + i\xi + \epsilon_{mn}) r_{mn}^\alpha r_{nm}^\beta E_\alpha^{\omega_1} E_\beta^{\omega_2}}{(\omega_1 - \epsilon_{mn} + i\xi)(\omega_2 - \epsilon_{nm} + i\xi)} \\ &= -\frac{1}{2} a_n^{(1;\alpha)} E_\alpha^{\omega_1},\end{aligned}\quad (37)$$

where we have used Eq.(20), Eq.(28), and $M_{mn}^{(1)*} = -M_{nm}^{(1)}$. We stress that $\mathbf{E} \cdot \mathbf{r}_c$ does not enter the semiclassical equations of motion since $\mathbf{r}_c$ and $\mathbf{k}_c$ are independent variables in the Lagrangian [4]. Note that Eq.(37) in the DC limit recovers the second-order energy correction obtained with the extended DC semiclassical theory [70, 71], as expected. Eqs.(34-35), combining with the Boltzmann equation, can be used to calculate the AC current density, as will be discussed in the following section.

III. CURRENT DENSITY

To clearly show the equivalence between the AC semiclassical theory and the response theory, in this section we first derive the current density (or the current density conductivity tensor) calculated from both the AC semiclassical theory formulated above and the response theory at the length gauge [43, 44], respectively.

A. The current density from the AC semiclassical theory

Armed with the AC semiclassical theory accurate up to the second order of the electric field, the current density

defined by [4]

$$J_\lambda = \sum_n \int_k \bar{f}_n \dot{r}_n^\lambda, \quad (38)$$

can be easily calculated by solving Eqs.(34-35) for the semiclassical velocity $\dot{r}_n^\lambda$ as well as Eq.(3) for the nonequilibrium distribution function $\bar{f}_n$. Particularly, up to the second order of the electric field, we find

$$\begin{aligned}\dot{r}_n^\lambda &= \left(v_n^\lambda + v_n^{(2;\lambda)} \right) + \left(\Omega_n^{\lambda\alpha} + \Omega_n^{(1;\lambda\alpha)} \right) E_\alpha^{\omega_1} \\ &\quad + \partial_t \left(a_n^{(1;\lambda)} + a_n^{(2;\lambda)} \right),\end{aligned}\quad (39)$$

where $v_n^{(2;\lambda)} = \partial_\lambda \epsilon_n^{(2)}$ with $\epsilon_n^{(2)}$ calculated in Eq.(37), $\Omega_n^{\lambda\alpha} = i \sum_m (r_{nm}^\lambda r_{mn}^\alpha - r_{nm}^\alpha r_{mn}^\lambda)$ is the conventional Berry curvature, and $\Omega_n^{(1;\lambda\alpha)} = \partial_\lambda a_n^{(1;\alpha)} - \partial_\alpha a_n^{(1;\lambda)}$ is the field-induced Berry curvature due to the first-order *positional shift* calculated in Eq.(28). In addition, by solving the Boltzmann equation we find (see Appendix B)

$$f_n^{(1)} = \frac{i\partial_\alpha f_n E_\alpha^{\omega_1}}{\omega_1 + i(\eta + \xi)}, \quad (40)$$

$$f_n^{(2)} = \frac{-\partial_{\alpha\beta}^2 f_n E_\alpha^{\omega_1} E_\beta^{\omega_2}}{[\omega_1 + i(\eta + \xi)][\omega_\Sigma + i(\eta + 2\xi)]}, \quad (41)$$

where $\eta \equiv 1/\tau$ is the scattering rate. Then by inserting Eqs.(39-41) into Eq.(38), we immediately obtain the first-order current density

$$J_\lambda^{(1)} \equiv J_\lambda^{(1;1)} + J_\lambda^{(1;0)} + J_\lambda^{(1;\bar{0})} \quad (42)$$

with

$$J_\lambda^{(1;1)} = \sum_n \int_k f_n^{(1)} v_n^\lambda = \sum_n \int_k \frac{i\partial_\alpha f_n v_n^\lambda E_\alpha^{\omega_1}}{\omega_1 + i(\eta + \xi)}, \quad (43)$$

$$J_\lambda^{(1;0)} = \sum_n \int_k f_n \Omega_n^{\lambda\alpha} E_\alpha^{\omega_1}, \quad (44)$$

$$J_\lambda^{(1;\bar{0})} = \sum_n \int_k f_n \partial_t a_n^{(1;\lambda)}, \quad (45)$$

where the bar over n indicates that the current density vanishes in the DC limit, and the second-order current density

$$J_\lambda^{(2)} \equiv J_\lambda^{(2;0)} + J_\lambda^{(2;1)} + J_\lambda^{(2;2)} \quad (46)$$

with

$$J_\lambda^{(2;0)} = \sum_n \int_k f_n \left(v_n^{(2;\lambda)} + \Omega_n^{(1;\lambda\alpha)} E_\alpha^{\omega_1} + \partial_t a_n^{(2;\lambda)} \right), \quad (47)$$

$$J_\lambda^{(2;1)} = \sum_n \int_k f_n^{(1)} \left(\Omega_n^{\lambda\alpha} E_\alpha^{\omega_1} + \partial_t a_n^{(1;\lambda)} \right), \quad (48)$$

$$\begin{aligned}J_\lambda^{(2;2)} &= \sum_n \int_k f_n^{(2)} v_n^\lambda \\ &= \sum_n \int_k \frac{-\partial_{\alpha\beta}^2 f_n v_n^\lambda E_\alpha^{\omega_1} E_\beta^{\omega_2}}{[\omega_1 + i(\eta + \xi)][\omega_\Sigma + i(\eta + 2\xi)]}.\end{aligned}\quad (49)$$

Note that both $J_\lambda^{(2;0)}$ and $J_\lambda^{(2;1)}$ contains contributions that disappear in the DC limit, which are defined as

$$J_\lambda^{(2;\bar{0})} \equiv \sum_n \int_k f_n \partial_t a_n^{(2;\lambda)}, \quad (50)$$

$$J_\lambda^{(2;\bar{1})} \equiv \sum_n \int_k f_n^{(1)} \partial_t a_n^{(1;\lambda)}. \quad (51)$$

In addition to the AC semiclassical theory, the current density can also be calculated by the response theory through solving the quantum Liouville equation for the nonequilibrium density matrix, which is summarized below.

B. The current density from the response theory

In subsection III A, we have expressed the results of AC semiclassical theory in terms of the current density; to make a clear notation distinction, below we will use the conductivity tensor of the current density to express the results given by the response theory. In particular, under the framework of the length-gauge response theory, we find that the linear conductivity can be decomposed as (see Appendix C)

$$\sigma_{\lambda\alpha}^{(1)} = \sigma_{\lambda\alpha}^{(1;1)} + \sigma_{\lambda\alpha}^{(1;0)} + \sigma_{\lambda\alpha}^{(1;\bar{0})}, \quad (52)$$

where

$$\sigma_{\lambda\alpha}^{(1;1)} = \sum_n \int_k \frac{i\partial_\alpha f_n}{\omega_1 + i\xi} v_n^\lambda, \quad (53)$$

$$\sigma_{\lambda\alpha}^{(1;0)} = \sum_n \int_k f_n \Omega_n^{\lambda\alpha}, \quad (54)$$

$$\sigma_{\lambda\alpha}^{(1;\bar{0})} = \sum_{mn} \int_k \frac{-i(\omega_1 + i\xi) r_{nm}^\lambda r_{mn}^\alpha f_{nm}}{\omega_1 - \epsilon_{mn} + i\xi}, \quad (55)$$

which contains three contributions same as the AC semiclassical theory.

Furthermore, in the second order of the electric field, the conductivity tensor evaluated with the response theory can be decomposed as (see Appendix C):

$$\sigma_{\lambda\alpha\beta}^{(2)} = \sigma_{\lambda\alpha\beta}^{(2,ee)} + \sigma_{\lambda\alpha\beta}^{(2,ei)} + \sigma_{\lambda\alpha\beta}^{(2,ie)} + \sigma_{\lambda\alpha\beta}^{(2,ii)}, \quad (56)$$

where

$$\sigma_{\lambda\alpha\beta}^{(2,ee)} = \sum_{mnl} \int_k \mathcal{V}_{nm}^\lambda \left(\frac{r_{ml}^\beta r_{ln}^\alpha f_{nl}}{\omega_1 - \epsilon_{ln} + i\xi} - \frac{f_{lm} r_{ml}^\alpha r_{ln}^\beta}{\omega_1 - \epsilon_{ml} + i\xi} \right), \quad (57)$$

$$\sigma_{\lambda\alpha\beta}^{(2,ei)} = \sum_{mn} \int_k \mathcal{V}_{nm}^\lambda \left(\frac{i r_{mn}^\beta \partial_\alpha f_{nm}}{\omega_1 + i\xi} \right), \quad (58)$$

$$\sigma_{\lambda\alpha\beta}^{(2,ie)} = \sum_{mn} \int_k \mathcal{V}_{nm}^\lambda \mathcal{D}_{mn}^\beta \left(\frac{i f_{nm} r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} \right), \quad (59)$$

$$\sigma_{\lambda\alpha\beta}^{(2,ii)} = \sum_n \int_k \frac{-v_n^\lambda \partial_{\alpha\beta}^2 f_n}{(\omega_1 + i\xi)(\omega_\Sigma + 2i\xi)}, \quad (60)$$

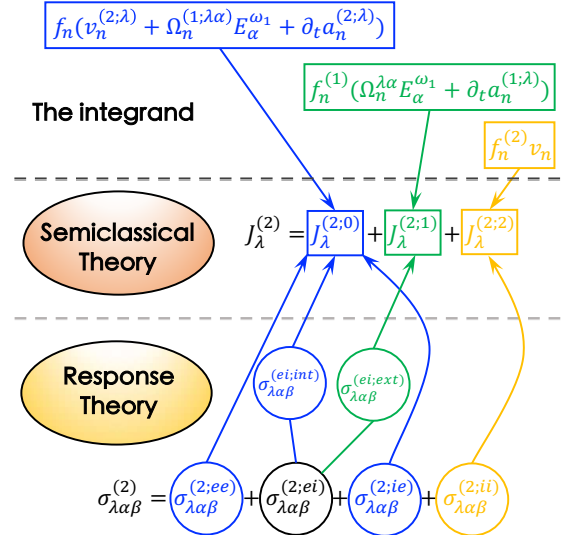


FIG. 1. The schematic illustration about the equivalence between the AC semiclassical theory and the response theory. Here the middle panel displays the second-order current density given by the AC semiclassical theory, where the integrand for each term is placed into the top panel, and the bottom panel shows the second-order nonlinear conductivity given by the length-gauge response theory. The arrows with different colors are used to indicate the equivalence between both formulations. We stress that $f_n \partial_t a_n^{2;\lambda}$ and $f_n^{(1)} \partial_t a_n^{1;\lambda}$ gives the intrinsic and extrinsic nonlinear displacement currents under an AC electric field, respectively, as firstly proposed in this work.

where $f_{nl} = f_n - f_l$, $\omega_\Sigma = \omega_1 + \omega_2$, and

$$\mathcal{V}_{nm}^\lambda = \frac{v_{nm}^\lambda}{\omega_\Sigma - \epsilon_{mn} + 2i\xi}. \quad (61)$$

Similar to the second-order current density calculated using the AC semiclassical theory, there exists many contributions at this order.

IV. EQUIVALENCE BETWEEN THE AC SEMICLASSICAL THEORY AND THE RESPONSE THEORY

Once the current density is obtained by both the AC semiclassical theory and the response theory, we are ready to show that the current density given by both formulations are equivalent. To that purpose, we note that the relaxation (indicated by τ or $\eta = 1/\tau$) has been explicitly included into the semiclassical currents (particularly in $J_\lambda^{(1;1)}$, $J_\lambda^{(2;1)}$ and $J_\lambda^{(2;2)}$) by way of solving the Boltzman equation and thereby corresponds to the extrinsic contributions, while the conductivities given by the response theory does not include any relaxation effects. However, by adopting the clean limit ($\tau \rightarrow \infty$ or $\eta \rightarrow 0$), we show that all these extrinsic semiclassical results can be exactly reproduced by the response theory,

in which they show the same $1/\xi$ -type divergent behavior under the DC limit ($\omega \rightarrow 0$) (Note that $\xi \rightarrow 0^+$). Recall that this divergence appeared in the semiclassical theory can be properly regulated by taking a finite τ due to the presence of relaxation, as a result, by replacing ξ with n/τ [63] in the corresponding expressions given by the response theory, we find that the equivalence between both formulations particularly in the clean limit can be inherited in the presence of relaxation. In addition to the extrinsic contributions, we further show that the intrinsic contributions (including $J_\lambda^{(1;0)}$ and $J_\lambda^{(2;0)}$ which are independent of τ) given by the semiclassical theory can also be exactly reproduced by the response theory, where $\xi \rightarrow 0$ can no longer lead to a divergent conductivity in the DC limit so that there is no need for regulating.

A. The first order

At the linear order, by comparing Eqs. (43-44) given by the AC semiclassical theory with Eqs. (53-54) given by the response theory, it is easy to find that

$$\sigma_{\lambda\alpha}^{(1;1)} E_\alpha^{\omega_1} = \lim_{\eta \rightarrow 0} J_\lambda^{(1;1)}, \quad \sigma_{\lambda\alpha}^{(1;0)} E_\alpha^{\omega_1} = J_\lambda^{(1;0)}, \quad (62)$$

which gives the well-known linear Drude current and intrinsic anomalous Hall current, respectively. Particularly, in the DC limit, we find

$$\begin{aligned} \lim_{\omega_1 \rightarrow 0} \lim_{\eta \rightarrow 0} J_\lambda^{(1;1)} &= \frac{1}{\xi} \sum_n \int_k \partial_\alpha f_n v_n^\lambda E_\alpha \\ &= \lim_{\omega_1 \rightarrow 0} \sigma_{\lambda\alpha}^{(1;1)} E_\alpha^{\omega_1}. \end{aligned} \quad (63)$$

Note that this divergence can be properly regulated by taking a finite τ particularly in the semiclassical result, as result, by replacing ξ by $1/\tau$ in the expression given by the response theory, we have

$$\begin{aligned} \lim_{\omega_1 \rightarrow 0} \lim_{\xi \rightarrow 0} J_\lambda^{(1;1)} &= \tau \sum_n \int_k \partial_\alpha f_n v_n^\lambda E_\alpha \\ &= \lim_{\omega_1 \rightarrow 0} \sigma_{\lambda\alpha}^{(1;1)} (\xi \rightarrow 1/\tau) E_\alpha^{\omega_1}. \end{aligned} \quad (64)$$

By doing this, we find that both formulations still give the same expression in the presence of relaxation. In addition, we have

$$\lim_{\omega_1 \rightarrow 0} J_\lambda^{(1;0)} = \sum_n \int_k f_n \Omega_n^{\lambda\alpha} E_\alpha = \lim_{\omega_1 \rightarrow 0} \sigma_{\lambda\alpha}^{(1;0)} E_\alpha^{\omega_1}, \quad (65)$$

which stands for the intrinsic contribution and therefore there is no need for regulating even in the DC limit.

Furthermore, by inserting Eq.(28) into Eq.(45), we find

$$\begin{aligned} J_\lambda^{(1;\bar{0})} &= \sum_{nm} \int_k f_n \left(\frac{r_{nm}^\lambda r_{mn}^\alpha \partial_t E_\alpha^{\omega_1}}{\omega_1 - \epsilon_{mn} + i\xi} + \frac{r_{mn}^\lambda r_{nm}^\alpha \partial_t E_\alpha^{-\omega_1}}{\omega_1 - \epsilon_{mn} - i\xi} \right) \\ &= \sum_{nm} \int_k f_{nm} \frac{-i(\omega_1 + i\xi) r_{nm}^\lambda r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} E_\alpha^{\omega_1} \\ &= \sigma_{\lambda\alpha}^{(1;\bar{0})} E_\alpha^{\omega_1}, \end{aligned} \quad (66)$$

where we have used $\sum_{\omega_1=\pm\omega} h(\omega_1) = \sum_{\omega_1=\pm\omega} h(-\omega_1)$ for an arbitrary function $h(\omega_1)$ and interchanged m and n to obtain the second line. We wish to mention that Eq.(45) or the equivalent $\sigma_{\lambda\alpha}^{(1;\bar{0})} E_\alpha^{\omega_1}$ gives the linear intrinsic displacement current under an AC electric field (vanishes in the DC limit), which is intimately related to the quantum metric as recently discussed in Ref. [62]. With Eqs. (62), (64), (65) and (66), we conclude that the AC semiclassical current at the linear order of the electric field is equivalent to that given by the response theory.

B. The second order: the DC case

Since the AC semiclassical theory and the response theory give many terms at the second order of the electric field, therefore, before considering the general situation we first show the equivalence between both formulations in the DC limit, where the contributions due to the time variation of *positional shift* in Eqs. (47) and (48) given by the AC semiclassical theory do not appear.

In the DC limit, by investigating the zero-frequency divergence of both approaches, we first note that Eq.(49) obtained with the AC semiclassical theory under the clean limit ($\eta \rightarrow 0$) is equivalent to Eq.(60) obtained with the response theory. Explicitly, we find

$$\begin{aligned} \lim_{\omega_i \rightarrow 0} \lim_{\eta \rightarrow 0} J_\lambda^{(2;2)} &= \frac{1}{\xi} \times \frac{1}{2\xi} \sum_n \int_k v_n^\lambda \partial_{\alpha\beta}^2 f_n E_\alpha E_\beta \\ &= \lim_{\omega_i \rightarrow 0} \sigma_{\lambda\alpha\beta}^{(2;ii)} E_\alpha^{\omega_1} E_\beta^{\omega_2}. \end{aligned} \quad (67)$$

Similarly, this divergence can also be properly regulated by taking a finite τ in the semiclassical theory, as result, by replacing $1/(2\xi^2)$ by τ^2 [63] in the expression given by the response theory, we arrive at

$$\begin{aligned} \lim_{\omega_i \rightarrow 0} \lim_{\xi \rightarrow 0} J_\lambda^{(2;2)} &= \tau^2 \sum_n \int_k v_n^\lambda \partial_{\alpha\beta}^2 f_n E_\alpha E_\beta \\ &= \lim_{\omega_i \rightarrow 0} \sigma_{\lambda\alpha\beta}^{(2;ii)} (2\xi^2 \rightarrow 1/\tau^2) E_\alpha^{\omega_1} E_\beta^{\omega_2}. \end{aligned} \quad (68)$$

By doing this, we find that both formulations still give the same result in the presence of relaxation, which is nothing but the familiar nonlinear Drude current which quadratically depends on the relaxation time τ .

Following the same strategy, we next show that the semiclassical current $J_\lambda^{(2;1)}$ given by Eq.(48) in the DC limit can be given by the nonlinear conductivity $\sigma_{\lambda\alpha\beta}^{(2,ei)}$ given by Eq.(58) in the response theory. Particularly, by inserting Eq.(40) into Eq.(48), we first find that $J_\lambda^{(2;1)}$ in the DC limit becomes

$$\lim_{\omega_i \rightarrow 0} \lim_{\eta \rightarrow 0} J_\lambda^{(2;1)} = \frac{1}{\xi} \sum_n \int_k \partial_\beta f_n \Omega_n^{\lambda\alpha} E_\alpha E_\beta, \quad (69)$$

On the other hand, using $v_{nm}^\lambda = i\epsilon_{nm}r_{nm}^\lambda$ ($m \neq n$) and

$$\begin{aligned} & \frac{\epsilon_{mn}}{(\omega_\Sigma - \epsilon_{mn} + 2i\xi)(\omega_1 + i\xi)} \\ &= \frac{\epsilon_{mn}}{\omega_2 - \epsilon_{mn} + i\xi} \left(\frac{1}{\omega_1 + i\xi} - \frac{1}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} \right), \quad (70) \end{aligned}$$

we find that Eq.(58) in the DC limit can be decomposed as $\sigma_{\lambda\alpha\beta}^{(2;ei)} = \sigma_{\lambda\alpha\beta}^{(ei;ext)} + \sigma_{\lambda\alpha\beta}^{(ei;int)}$, where

$$\lim_{\omega_i \rightarrow 0} \sigma_{\lambda\alpha\beta}^{(ei;ext)} = \frac{1}{\xi} \sum_{mn} \int_k i r_{nm}^\lambda r_{mn}^\beta \partial_\alpha f_{nm}, \quad (71)$$

$$\lim_{\omega_i \rightarrow 0} \sigma_{\lambda\alpha\beta}^{(ei;int)} = \sum_{mn} \int_k \frac{r_{nm}^\lambda r_{mn}^\beta \partial_\alpha f_{nm}}{-\epsilon_{mn}}. \quad (72)$$

Here the superscript *ext* (*int*) stands for the extrinsic (intrinsic) contribution. By interchanging m and n of Eq.(71), it is easy to show that

$$\begin{aligned} \lim_{\omega_i \rightarrow 0} \sigma_{\lambda\alpha\beta}^{(ei;ext)} E_\alpha^{\omega_1} E_\beta^{\omega_2} &= \frac{1}{\xi} \sum_n \int_k \partial_\alpha f_n \Omega_n^{\lambda\beta} E_\alpha E_\beta \\ &= \lim_{\omega_i \rightarrow 0} \lim_{\eta \rightarrow 0} J_\lambda^{(2;1)}. \quad (73) \end{aligned}$$

Beyond the clean limit or when the relaxation is present, this divergent conductivity can be naturally regulated by taking a finite τ in the semiclassical expression, as a result, also by replacing $1/\xi$ by τ , we have

$$\begin{aligned} \lim_{\omega_i \rightarrow 0} \sigma_{\lambda\alpha\beta}^{(ei;ext)} (\xi \rightarrow 1/\tau) E_\alpha^{\omega_1} E_\beta^{\omega_2} &= \tau \sum_n \int_k \partial_\alpha f_n \Omega_n^{\lambda\beta} E_\alpha E_\beta \\ &= \lim_{\omega_i \rightarrow 0} \lim_{\xi \rightarrow 0} J_\lambda^{(2;1)}, \quad (74) \end{aligned}$$

which is nothing but the well-known extrinsic second-order nonlinear Hall current driven by the Berry curvature dipole [6, 90].

Finally, we show that the intrinsic conductivity Eq.(72) together with the remaining conductivities (which can only contribute to the intrinsic current, as will be clear below) in the response theory, namely, Eq.(57) and Eq.(59), is equivalent to the intrinsic current given by Eq.(47) in the AC semiclassical theory, particularly in the DC limit. Explicitly, we will show:

$$\lim_{\omega_i \rightarrow 0} J_\lambda^{(2;0)} = \lim_{\omega_i \rightarrow 0} \left[\sigma_{\lambda\alpha\beta}^{(2;ee)} + \sigma_{\lambda\alpha\beta}^{(2;ie)} + \sigma_{\lambda\alpha\beta}^{(ei;int)} \right] E_\alpha^{\omega_1} E_\beta^{\omega_2}. \quad (75)$$

To that purpose, by inserting Eq.(37) and $\Omega_n^{(1;\lambda\alpha)} = \partial_\lambda a_n^{(1;\alpha)} - \partial_\alpha a_n^{(1;\lambda)}$ into Eq.(47), we find that $J_\lambda^{(2;0)}$ given by the AC semiclassical theory in the DC limit can be

simplified as

$$\begin{aligned} & \lim_{\omega_i \rightarrow 0} J_\lambda^{(2;0)} \\ &= \sum_n \int_k f_n \left[-\frac{\partial_\lambda a_n^{(1;\beta)}}{2} + \left(\partial_\lambda a_n^{(1;\beta)} - \partial_\beta a_n^{(1;\lambda)} \right) \right] E_\beta \\ &= \sum_n \int_k \left[a_n^{(1;\lambda)} \partial_\beta f_n - \frac{1}{2} a_n^{(1;\beta)} \partial_\lambda f_n \right] E_\beta \\ &= \sum_{nm} \int_k \left(\frac{g_{nm}^{\lambda\alpha}}{\epsilon_{nm}} \partial_\beta f_n - \frac{g_{nm}^{\beta\alpha}}{2\epsilon_{nm}} \partial_\lambda f_n \right) E_\alpha E_\beta \\ &= \frac{1}{2} \sum_{nm} \int_k \left(\frac{g_{nm}^{\lambda\alpha}}{\epsilon_{nm}} \partial_\beta f_n + \frac{g_{nm}^{\lambda\beta}}{\epsilon_{nm}} \partial_\alpha f_n - \frac{g_{nm}^{\beta\alpha}}{\epsilon_{nm}} \partial_\lambda f_n \right) E_\alpha E_\beta, \quad (76) \end{aligned}$$

where we have symmetrized the final result about the field indices. Note that in the DC limit $a_n^{(1;\lambda)} = \sum_m g_{nm}^{\lambda\alpha} E_\alpha / \epsilon_{nm}$ with $g_{nm}^{\lambda\alpha} = r_{nm}^\lambda r_{mn}^\alpha + r_{nm}^\alpha r_{mn}^\lambda$ the quantum metric, which is symmetric about λ and α .

On the other hand, by interchanging m and n and symmetrizing α and β , we find that Eq.(72) can be expressed as

$$\sigma_{\lambda\alpha\beta}^{(ei;int)} = \sum_{mn} \int_k \frac{1}{2} \left(\frac{g_{nm}^{\lambda\beta}}{\epsilon_{nm}} \partial_\alpha f_n + \frac{g_{nm}^{\lambda\alpha}}{\epsilon_{nm}} \partial_\beta f_n \right). \quad (77)$$

In addition, for $\sigma_{\lambda\alpha\beta}^{(2;ie)}$, in the DC limit we find

$$\lim_{\omega_i \rightarrow 0} \sigma_{\lambda\alpha\beta}^{(2;ie)} = \sum_{mn} \int_k \frac{f_{nm} r_{mn}^\alpha}{\epsilon_{nm}} \mathcal{D}_{nm}^\beta r_{nm}^\lambda. \quad (78)$$

Furthermore, for $\sigma_{\lambda\alpha\beta}^{(2;ee)}$, when $m = n$ we find (see Appendix D 1)

$$\begin{aligned} \sigma_{\lambda\alpha\beta}^{(2;ee)} (m = n) &\equiv \sigma_{\lambda\alpha\beta}^{(2;ee1)} \\ &= \frac{1}{2} \sum_{nm} \int_k \frac{\Delta_{nm}^\lambda f_{nm} r_{nm}^\beta r_{mn}^\alpha}{(\omega_1 - \epsilon_{mn} + i\xi)(\omega_2 + \epsilon_{mn} + i\xi)}, \quad (79) \end{aligned}$$

where $\Delta_{nm}^\lambda = v_n^\lambda - v_m^\lambda$. As shown in Appendix D 1, the divergent factor $1/(\omega_\Sigma + i2\xi)$ in $\sigma_{\lambda\alpha\beta}^{(2;ee)} (m = n)$ has been removed after symmetrization. In the DC limit, we then obtain

$$\begin{aligned} \lim_{\omega_i \rightarrow 0} \sigma_{\lambda\alpha\beta}^{(2;ee1)} &\equiv \frac{1}{2} \sum_{nm} \int_k \frac{-\Delta_{nm}^\lambda f_{nm} r_{nm}^\beta r_{mn}^\alpha}{\epsilon_{nm}^2} \\ &= \frac{1}{2} \sum_{nm} \int_k f_n g_{nm}^{\beta\alpha} \partial_\lambda \left(\frac{1}{\epsilon_{nm}} \right), \quad (80) \end{aligned}$$

which is symmetric about the field indices α and β . When $m \neq n$, we can directly take the DC limit of $\sigma_{\lambda\alpha\beta}^{(2;ee)}$ and

obtain

$$\lim_{\omega_i \rightarrow 0} \sigma_{\lambda\alpha\beta}^{(2;ee)} (m \neq n) \equiv \lim_{\omega_i \rightarrow 0} \sigma_{\lambda\alpha\beta}^{(2;ee2)} \quad (81)$$

$$\begin{aligned} &= \sum_{nml} \int_k i r_{nm}^\lambda \left(\frac{r_{ml}^\beta r_{ln}^\alpha f_{nl}}{-\epsilon_{ln}} - \frac{f_{lm} r_{ml}^\alpha r_{ln}^\beta}{-\epsilon_{ml}} \right) \\ &= \sum_{nml} \int_k \frac{r_{mn}^\alpha f_{nm}}{\epsilon_{nm}} i \left(r_{nl}^\lambda r_{lm}^\beta - r_{nl}^\beta r_{lm}^\lambda \right) \\ &= \sum_{nm} \int_k \frac{r_{mn}^\alpha f_{nm}}{\epsilon_{nm}} (\mathcal{D}_{nm}^\lambda r_{nm}^\beta - \mathcal{D}_{nm}^\beta r_{nm}^\lambda), \end{aligned} \quad (82)$$

where we have used the relation [44]

$$\mathcal{D}_{nm}^\lambda r_{nm}^\beta - \mathcal{D}_{nm}^\beta r_{nm}^\lambda = i \sum_l \left(r_{nl}^\lambda r_{lm}^\beta - r_{nl}^\beta r_{lm}^\lambda \right). \quad (83)$$

Combining Eq.(78), Eq.(80) and Eq.(82), we find

$$\begin{aligned} &\lim_{\omega_i \rightarrow 0} \left(\sigma_{\lambda\alpha\beta}^{(2;ie)} + \sigma_{\lambda\alpha\beta}^{(2;ee)} \right) \equiv \sigma_{\lambda\alpha\beta}^{(2;ie)} + \sigma_{\lambda\alpha\beta}^{(2;ee1)} + \sigma_{\lambda\alpha\beta}^{(2;ee2)} \\ &= \sum_{nm} \int_k \left[\frac{r_{mn}^\alpha f_{nm}}{\epsilon_{nm}} \mathcal{D}_{nm}^\lambda r_{nm}^\beta + \frac{1}{2} f_n g_{nm}^{\beta\alpha} \partial_\lambda \left(\frac{1}{\epsilon_{nm}} \right) \right] \\ &= \frac{1}{2} \sum_{nm} \int_k f_n \left[\frac{1}{\epsilon_{nm}} \partial_\lambda g_{nm}^{\beta\alpha} + g_{nm}^{\beta\alpha} \partial_\lambda \left(\frac{1}{\epsilon_{nm}} \right) \right] \\ &= -\frac{1}{2} \sum_{nm} \int_k \frac{g_{nm}^{\beta\alpha}}{\epsilon_{nm}} \partial_\lambda f_n, \end{aligned} \quad (84)$$

where we have symmetrized the first term of the second line about α and β to obtain the final result and used the following relation [44]

$$\partial_\lambda (r_{nm}^\alpha r_{mn}^\beta) = (\mathcal{D}_{nm}^\lambda r_{nm}^\alpha) r_{mn}^\beta + r_{nm}^\alpha (\mathcal{D}_{mn}^\lambda r_{mn}^\beta). \quad (85)$$

By further combining Eq.(84) with Eq.(77), we finally arrive at

$$\begin{aligned} &\lim_{\omega_i \rightarrow 0} \left[\sigma_{\lambda\alpha\beta}^{(2;ie)} + \sigma_{\lambda\alpha\beta}^{(2;ee)} + \sigma_{\lambda\alpha\beta}^{(ei;int)} \right] E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ &= \frac{1}{2} \sum_{nm} \int_k \left(\frac{g_{nm}^{\lambda\alpha}}{\epsilon_{nm}} \partial_\beta f_n + \frac{g_{nm}^{\lambda\beta}}{\epsilon_{nm}} \partial_\alpha f_n - \frac{g_{nm}^{\beta\alpha}}{\epsilon_{nm}} \partial_\lambda f_n \right) E_\alpha E_\beta \\ &= \lim_{\omega_i \rightarrow 0} J_\lambda^{(2;0)}, \end{aligned} \quad (86)$$

which shows that the intrinsic second-order nonlinear current obtained with the response theory is also equivalent to that derived with the AC semiclassical theory, particularly in the DC limit. Note that this intrinsic second-order nonlinear current contains both the longitudinal (from the second-order energy correction $\epsilon_n^{(2)}$) and transverse (from both the anomalous velocity $\mathbf{E} \times \boldsymbol{\Omega}_n^{(1)}$ and the second-order energy correction $\epsilon_n^{(2)}$) components, which can only appear in systems without $\mathcal{P}$ ($\mathcal{P}$, inversion) and $\mathcal{T}$ symmetries.

At this stage, we conclude that the intrinsic second-order Ohmic current proposed in Refs. [65, 66] based on the response theory can also be reproduced by the

semiclassical theory when the second-order wavepacket energy is taken into account (ignored in Ref. [65]). As a result, the response theory does not contain any inter-band coherent correction compared to the semiclassical theory. We stress that the intrinsic contributions given by both formulations are irrelevant to the relaxation so that there is no need for regulating ($\xi \rightarrow 0$ does not lead to a divergence) even in the DC limit. In other words, the semiclassical theory give the same intrinsic physics as the response theory. Finally, to clearly illustrate the equivalence between two formulations, we schematically summarized the main results obtained in this subsection into FIG.(1), which will be useful when we show that both formulations are equivalent in the AC regime in the following.

C. The second order: the AC case

Guided by the equivalence between the semiclassical theory and the response theory in the DC limit, as summarized in FIG.(1), we further show the equivalence between both approaches in the AC regime. First of all, it is easy to show that

$$\sigma_{\lambda\alpha\beta}^{(2;ii)} E_\alpha^{\omega_1} E_\beta^{\omega_2} = \lim_{\eta \rightarrow 0} J_\lambda^{(2;2)} \quad (87)$$

by using Eq.(60) and Eq.(49), in which Eq.(41) is inserted.

In the DC case, we have shown that the conductivity $\sigma_{\lambda\alpha\beta}^{(2;ei)}$ can be divided into the extrinsic and intrinsic contributions. In the AC case, we follow the same partition. In addition, we may expect some additional contributions that vanish in the DC limit, which should correspond to the displacement current given by the time variation of *positional shift* in the AC semiclassical theory. Particularly, using Eq.(70) and

$$\frac{\epsilon_{mn}}{\omega_2 - \epsilon_{mn} + i\xi} = -1 + \frac{\omega_2 + i\xi}{\omega_2 - \epsilon_{mn} + i\xi}, \quad (88)$$

we can partition $\sigma_{\lambda\alpha\beta}^{(2;ei)}$ into $\sigma_{\lambda\alpha\beta}^{(2;ei)} \equiv \sigma_{\lambda\alpha\beta}^{(ei;1)} + \sigma_{\lambda\alpha\beta}^{(ei;0)} + \sigma_{\lambda\alpha\beta}^{(ei;\bar{1})}$, where

$$\sigma_{\lambda\alpha\beta}^{(ei;1)} = \sum_{mn} \int_k \frac{-r_{nm}^\lambda r_{mn}^\beta \partial_\alpha f_{nm}}{\omega_1 + i\xi}, \quad (89)$$

$$\sigma_{\lambda\alpha\beta}^{(ei;\bar{1})} = \sum_{mn} \int_k \frac{(\omega_2 + i\xi) r_{nm}^\lambda r_{mn}^\beta \partial_\alpha f_{nm}}{(\omega_2 - \epsilon_{mn} + i\xi)(\omega_1 + i\xi)}, \quad (90)$$

$$\begin{aligned} \sigma_{\lambda\alpha\beta}^{(ei;0)} &\equiv \sum_{mn} \int_k \frac{-\epsilon_{mn} r_{nm}^\lambda r_{mn}^\beta \partial_\alpha f_{nm}}{(\omega_2 - \epsilon_{mn} + i\xi)(\omega_\Sigma - \epsilon_{mn} + i2\xi)} \\ &= \sum_{mn} \int_k \frac{-i v_{nm}^\lambda r_{mn}^\alpha \partial_\beta f_{nm}}{(\omega_1 - \epsilon_{mn} + i\xi)(\omega_\Sigma - \epsilon_{mn} + i2\xi)}. \end{aligned} \quad (91)$$

Here the superscripts $(ei;1)$, $(ei;\bar{1})$, and $(ei;0)$ stand for the extrinsic (existing in the DC limit), AC (disappearing

in the DC limit), and intrinsic (existing in the DC limit) contributions, respectively. As expected, we find

$$\left(\sigma_{\lambda\alpha\beta}^{(ei;1)} + \sigma_{\lambda\alpha\beta}^{(ei;\bar{1})}\right) E_{\alpha}^{\omega_1} E_{\beta}^{\omega_2} = \lim_{\eta \rightarrow 0} J_{\lambda}^{(2;1)}, \quad (92)$$

where

$$\begin{aligned} \lim_{\eta \rightarrow 0} J_{\lambda}^{(2;1)} &= \lim_{\eta \rightarrow 0} \sum_n \int_k f_n^{(1)} \left(\Omega_n^{\lambda\beta} E_{\beta}^{\omega_2} + \partial_t a_n^{(1;\lambda)} \right) \\ &= \sum_n \int_k \frac{-r_{nm}^{\lambda} r_{mn}^{\beta} \partial_{\alpha} f_{nm}}{\omega_1 + i\xi} E_{\alpha}^{\omega_1} E_{\beta}^{\omega_2} \\ &+ \sum_{mn} \int_k \frac{(\omega_2 + i\xi) r_{nm}^{\lambda} r_{mn}^{\beta} \partial_{\alpha} f_{nm}}{(\omega_2 - \epsilon_{mn} + i\xi)(\omega_1 + i\xi)} E_{\alpha}^{\omega_1} E_{\beta}^{\omega_2} \end{aligned} \quad (93)$$

by inserting Eq.(28) and Eq.(40) into Eq.(48). Note that $\Omega_n^{\lambda\beta} = \sum_m i(r_{nm}^{\lambda} r_{mn}^{\beta} - r_{nm}^{\beta} r_{mn}^{\lambda})$ and the second line is obtained by index interchanging.

For the remaining terms both in the response theory and in the AC semiclassical theory, we can also establish the following equivalent relation (see Appendix D 2)

$$\left(\sigma_{\lambda\alpha\beta}^{(2;ee)} + \sigma_{\lambda\alpha\beta}^{(2;ie)} + \sigma_{\lambda\alpha\beta}^{(ei;0)}\right) E_{\alpha}^{\omega_1} E_{\beta}^{\omega_2} = J_{\lambda}^{(2;0)}, \quad (94)$$

which is the same as the DC case, as illustrated in FIG.(1).

As expected, the AC semiclassical theory is also equivalent to the response theory at the second order of the electric field, particularly in the clean limit $\eta \rightarrow 0$. Beyond this limit, we can still introduce the relaxation in the response theory in the same way as we have adopted in the DC limit. By doing that, the semiclassical results at both the zero and finite frequencies can be reproduced by the response theory. Besides this manual way based on the equivalence between both formulations, we wish to remark that the previous relaxation schemes adopted in the response theory particularly by modifying the quantum Liouville equation in fact is unreasonable, as will be explained in section V C.

V. CONSEQUENCES OF THE EQUIVALENCE

In this section, we discuss the implications behind the equivalence between the AC semiclassical theory and the response theory. First, we show that the intrinsic nonlinear Ohmic current and the nonreciprocal magnetoresistance represent the same physics. Second, we suggest that the energy used in the equilibrium Fermi distribution in the semiclassical theory is the unperturbed band energy by assuming that both formulations give the same intrinsic responses. Third, we argue that by phenomenally modifying the quantum Liouville equation to introduce relaxation in the response theory needs to be reconsidered.

A. Intrinsic nonlinear Ohmic current versus nonreciprocal magnetoresistance

In previous section, we have shown that the intrinsic nonlinear Ohmic current, as first derived with the response theory [65, 66], can be obtained by the semiclassical theory with the consideration of the second-order wavepacket energy correction, which has been suppressed in the pioneering paper Ref. [7] when evaluating the intrinsic second-order nonlinear current. Interestingly, rather than using the wavepacket method, we note that a second-order energy calculated with the Kohn-Luttinger method [69], can also lead to an intrinsic nonlinear longitudinal current (nonreciprocal magnetoresistance [69]), which has been added to the extended second-order semiclassical results given by Ref. [7] due to the same quantum metric origin. And this second-order energy correction also plays a key role in understanding the gravitational anomaly [86].

Unfortunately, we wish to point out that the energy calculated with the Kohn-Luttinger method can not be directly used in the semiclassical theory. The wavepacket energy in the semiclassical theory accurate up to the second order of the electric field is given by [70, 71]

$$\bar{\epsilon}_n = \epsilon_n + \mathbf{E} \cdot \mathbf{r}_c - \frac{1}{2} \sum_m \frac{g_{nm}^{\alpha\beta} E_{\alpha} E_{\beta}}{\epsilon_{nm}}, \quad (95)$$

where we take the DC limit and

$$\mathbf{r}_c = \partial_{\mathbf{k}_c} \gamma + \mathbf{A}_n + \mathbf{a}_n^{(1)} + \mathbf{a}_n^{(2)} \dots \quad (96)$$

As stated previously, $\mathbf{E} \cdot \mathbf{r}_c$ does not enter the semiclassical equations of motion since $\mathbf{r}_c$ and $\mathbf{k}_c$ are independent variables in the Lagrangian [4]. Explicitly, the energy appeared in the semiclassical equations of motion is [70, 71]

$$\bar{\epsilon}_n^{\text{EOM}} = \epsilon_n - \frac{1}{2} \sum_m \frac{g_{nm}^{\alpha\beta} E_{\alpha} E_{\beta}}{\epsilon_{nm}}. \quad (97)$$

On the other hand, the energy calculated with the Kohn-Luttinger method is given by

$$\bar{\epsilon}_n^{\text{KL}} = \epsilon_n + \frac{1}{2} \sum_m \frac{g_{nm}^{\alpha\beta} E_{\alpha} E_{\beta}}{\epsilon_{nm}}, \quad (98)$$

which is reproduced in Appendix E by exactly following Ref. [69]. Compared to the semiclassical wavepacket energy $\bar{\epsilon}_n^{\text{EOM}}$, we note that the second-order energy in $\bar{\epsilon}_n^{\text{KL}}$ differs a sign with that in $\bar{\epsilon}_n^{\text{EOM}}$. Notably, we note that $\bar{\epsilon}_n^{\text{KL}}$ with the Kohn-Luttinger method bypasses the gauge-variant energy correction $\mathbf{E} \cdot \mathbf{A}_n$ by choosing a special gauge. And it is this gauge choice that leads to the inconsistency between $\bar{\epsilon}_n^{\text{EOM}}$ and $\bar{\epsilon}_n^{\text{KL}}$ since the gauge-variant energy $\mathbf{E} \cdot \mathbf{r}_c$ bypassed in the semiclassical theory also contains a second-order correction

$$\Delta \bar{\epsilon}_n^{(2)} = \mathbf{E} \cdot \mathbf{a}_n^{(1)} = \sum_n \frac{g_{nm}^{\alpha\beta} E_{\alpha} E_{\beta}}{\epsilon_{nm}}. \quad (99)$$

By removing this correction from $\bar{\epsilon}_n^{\text{KL}}$, we find [82]:

$$\bar{\epsilon}_n^{\text{KL}} - \Delta \bar{\epsilon}_n^{(2)} = \epsilon_n - \frac{1}{2} \sum_m \frac{g_{nm}^{\alpha\beta} E_\alpha E_\beta}{\epsilon_{nm}} = \bar{\epsilon}_n^{\text{EOM}}. \quad (100)$$

In section IV, we have shown that the second-order wavepacket energy in the semiclassical theory is responsible for the intrinsic nonlinear Ohmic current, as firstly derived by the response theory. As a result, the nonreciprocal magnetoresistance (intrinsic nonlinear longitudinal current) that arises from the same energy correction in fact represents the same physics with the intrinsic nonlinear Ohmic current. Particularly, by using Eq.(76) and defining $\lim_{\omega_i \rightarrow 0} J_\lambda^{(2;0)} \equiv \sigma_{\alpha\beta}^\lambda E_\alpha E_\beta$, we find

$$\sigma_{\alpha\beta}^\lambda = \frac{1}{2} \sum_{nm} \int_k \left(\frac{g_{nm}^{\lambda\alpha}}{\epsilon_{nm}} \partial_\beta f_n + \frac{g_{nm}^{\lambda\beta}}{\epsilon_{nm}} \partial_\alpha f_n - \frac{g_{nm}^{\beta\alpha}}{\epsilon_{nm}} \partial_\lambda f_n \right),$$

which contains both the intrinsic nonlinear Hall and Ohmic (or nonreciprocal magnetoresistance) currents.

B. Energy used in the distribution function

Note that in the previous sections, we have assumed that the energy used in the equilibrium Fermi distribution function is the unperturbed band energy. Recently, some works obtained the third-order nonlinear Hall effects [9, 87–89] by Taylor expanding the Fermi distribution function where the energy argument of the equilibrium Fermi distribution function takes into account the correction due to the external field. For instance, by replacing the unperturbed energy in $f_n^{(1)}$ with $\bar{\epsilon}_n = \epsilon_n + \epsilon_n^{(2)}$ and further Taylor expanding $f_n^{(1)}(\bar{\epsilon}_n)$ as

$$\begin{aligned} f_n^{(1)}(\bar{\epsilon}_n) &= \tau E_\alpha \partial_\alpha f_n(\bar{\epsilon}_n) \\ &= \tau E_\alpha \partial_\alpha f_n(\epsilon_n) + \tau E_\alpha \partial_\alpha \left[\epsilon_n^{(2)} f_n'(\epsilon_n) \right], \end{aligned} \quad (101)$$

one can immediately obtain an extrinsic third-order nonlinear current

$$J_\lambda = \tau \sum_n \int_k v_n^\lambda E_\alpha \partial_\alpha \left[\epsilon_n^{(2)} f_n'(\epsilon_n) \right]. \quad (102)$$

Note that Eq.(101) taken the DC limit of Eq.(40). However, based on the equivalence between both formulations, we argue that these third-order extrinsic current responses can not appear.

In particular, if the energy argument used in the equilibrium Fermi distribution function considers the correction under the electric field, it is easy to show that [7, 91]:

$$\int_k f_n(\bar{\epsilon}_n) \bar{\mathbf{v}}_n^a = \int_k f_n(\bar{\epsilon}_n) \frac{\partial \bar{\epsilon}_n}{\partial \mathbf{k}} = \int_k \frac{\partial g_n(\bar{\epsilon}_n)}{\partial \mathbf{k}} = 0, \quad (103)$$

where $g_n = -k_B T \ln [1 + e^{-(\bar{\epsilon}_n - \mu)/k_B T}]$ [71] and the band summation over n is suppressed for brevity. Clearly,

Eq.(103) forbids the intrinsic nonlinear longitudinal current arising from the field-induced group velocity, but these intrinsic nonlinear longitudinal current can be allowed by the response theory. As a result, by assuming that both formulations give the same intrinsic physics, we suggest that the energy used in the equilibrium Fermi distribution in the semiclassical theory is the unperturbed band energy, as we have adopted in section III and section IV.

C. Relaxation in the response theory

Finally, we discuss the phenomenological relaxation introduced in the response theory. We mention that the conductivity tensors Eqs.(57-60) from the response theory are obtained by solving the quantum Liouville equation Eq.(4) and hence do not contain relaxation like the semiclassical theory, where the relaxation is explicitly introduced through solving the Boltzmann equation. Usually, to take the relaxation into account in the response theory, Eq.(4) is phenomenologically modified as [72, 73]

$$i\partial_t \rho_{mn} = [H, \rho]_{mn} - \frac{i}{\tau} (\rho_{mn} - \rho_{mn}^{(0)}), \quad (104)$$

which has been used in many studies [74–76] to evaluate the nonlinear responses. Interestingly, we find that this phenomenological modification will give (see Appendix C3),

$$\sigma_{\lambda\alpha}^H = 2 \sum_{mn} \int_k f_n \text{Re} \left[\frac{r_{mn}^\alpha v_{nm}^\lambda}{-\epsilon_{mn} + i/\tau} \right]. \quad (105)$$

We have the following observations about this result. First, Eq. (105) can deliver a linear extrinsic Hall current in $\mathcal{T}$ -invariant systems (Note that $\sigma_{\lambda\alpha}^H$ is a $\mathcal{T}$ -even rank-2 tensor), which contradicts with the current experimental recognition and in fact is also not allowed by the Onsager reciprocal relation [12, 28, 83]. Second, Eq. (105) can give a τ^{-1} -dependent current responses when $\hbar/\tau \gg \epsilon_{mn}$, which can not show up in the semiclassical results but surprisingly appear in the nonlinear current responses [64] based on the response theory. Third, the equivalence between both formulations is broken. Therefore, we argue that the additional term in Eq.(104) is unreasonable.

Interestingly, if the phenomenological relaxation introduced in Eq.(104) is constrained to the intraband scattering, namely by modifying Eq.(104) into

$$i\partial_t \rho_{mn} = [H, \rho]_{mn} - \frac{i}{\tau} \delta_{mn} (\rho_{mn} - \rho_{mn}^{(0)}), \quad (106)$$

we find that the response theory agrees perfectly with the semiclassical theory at the linear order of the electric field, as shown in Appendix C3. Furthermore, at the second order of the electric field, Eq.(106) gives (see

Appendix C3),

$$\sigma_{\lambda\alpha\beta}^{(2,ee)} = \sum_{mnl} \int_k \bar{V}_{nm}^\lambda \left(\frac{r_{ml}^\beta r_{ln}^\alpha f_{nl}}{\omega_1 - \epsilon_{ln} + i\xi} - \frac{f_{lm} r_{ml}^\alpha r_{ln}^\beta}{\omega_1 - \epsilon_{ml} + i\xi} \right), \quad (107)$$

$$\sigma_{\lambda\alpha\beta}^{(2,ei)} = \sum_{mn} \int_k \bar{V}_{nm}^\lambda \left(\frac{i r_{mn}^\beta \partial_\alpha f_{nm}}{\omega_1 + i(\eta + \xi)} \right), \quad (108)$$

$$\sigma_{\lambda\alpha\beta}^{(2,ie)} = \sum_{mn} \int_k \bar{V}_{nm}^\lambda \mathcal{D}_\beta^{mn} \left(\frac{i f_{nm} r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} \right), \quad (109)$$

$$\sigma_{\lambda\alpha\beta}^{(2,ii)} = \sum_n \int_k \frac{-v_n^\lambda \partial_{\alpha\beta}^2 f_n}{[\omega_1 + i(\eta + \xi)] [\omega_\Sigma + i(\eta + 2\xi)]}, \quad (110)$$

where $\bar{\eta} = \eta \delta_{mn}$ (note that $\eta = 1/\tau$) and

$$\bar{V}_{nm}^\lambda \equiv \frac{v_{nm}^\lambda}{\omega_\Sigma - \epsilon_{mn} + i(\bar{\eta} + 2\xi)}. \quad (111)$$

At this stage, we first note that Eqs. (110) and (108), respectively, can also perfectly reproduce the extrinsic nonlinear Drude and Hall currents given by Eqs. (49) and (48) in the AC semiclassical theory, as outlined in FIG. 1. Note that that Eq. (110) also contains the intrinsic contribution, see FIG. 1. Unfortunately, we find that Eq. (107) particularly with $m = n$ can no longer give an intrinsic current like Eq. (80) given by the original quantum Liouville equation, although the intrinsic currents contributed by Eqs. (108) and (109) keep unchanged. Therefore, the intrinsic current given by the semiclassical theory can no longer be reproduced by the modified response theory, particularly in terms of Eq. (106).

In fact, in section IV, a scheme to include relaxation in the response theory has been utilized, particularly by regulating the clean zero-frequency divergent conductivities given by both formulations on the same footing. However, we can not rule out other ways of introducing the relaxation as long as it will not lead to unphysical results. To close this section, we believe that the distinction between the semiclassical theory and the response theory is rooted in the different ways of including the relaxation. And we conclude that the previous scheme to include relaxation in the response theory by modifying the Liouville equation for the nonequilibrium density matrix needs to be reconsidered.

VI. AC NONLINEAR EFFECTS

After establishing the equivalence between both formulations, in this section we focus on the overlooked second-order nonlinear current responses unique to the AC transport, which contains the intrinsic second-order nonlinear displacement current from the time variation of the second-order *positional shift* and the extrinsic second-order nonlinear displacement current, which is obtained

by combining the first-order nonequilibrium Fermi distribution with the time variation of the first-order *positional shift*. Although both formulations can give these two contributions, we will use the expressions given by the AC semiclassical theory since it gives a clear partition and presents a clear physical origin of the *positional shift*.

A. The intrinsic nonlinear displacement current

From the extended AC semiclassical theory, the intrinsic nonlinear displacement current is given by

$$J_\lambda^{(2;\bar{0})} = \sum_n \int_k f_n \partial_t a_n^{(2;\lambda)} = -\omega \sigma_{\alpha\beta}^\lambda E_\alpha E_\beta^\omega, \quad (112)$$

where E_α and $E_\beta^\omega = E_\beta \sin(\omega t)$ stands for the applied DC and AC electric fields, respectively. Under this crossed setup, we find

$$\begin{aligned} \sigma_{\alpha\beta}^\lambda = & \sum_{nml} \int_k f_n \text{Re} \left(\frac{2r_{nm}^\lambda r_{ml}^\beta r_{ln}^\alpha + r_{nl}^\beta r_{lm}^\lambda r_{mn}^\alpha}{\epsilon_{nl} \epsilon_{nm}} \right) \\ & + \sum_{nm} \int_k f_n \text{Re} \left[i \mathcal{D}_{mn}^\beta \left(\frac{r_{mn}^\alpha}{\epsilon_{nm}} \right) \frac{2r_{nm}^\lambda}{\epsilon_{nm}} \right] \\ & - \sum_{nm} \int_k f_n \text{Re} \left[i \mathcal{D}_{nm}^\lambda \left(\frac{r_{nm}^\alpha}{\epsilon_{nm}} \right) \frac{r_{mn}^\beta}{\epsilon_{nm}} \right], \end{aligned} \quad (113)$$

which is obtained by taking $\omega_1 = 0$ and summing over $\omega_2 = \pm\omega$ in Eq. (29). In addition, $\hbar\omega \ll \epsilon_{mn}$ is assumed.

The same as the linear intrinsic displacement current [62], we note that $\sigma_{\alpha\beta}^\gamma$ also features the $\mathcal{T}$ -even ($\mathcal{T}$, time reversal) behavior, which indicates that this nonlinear current can appear in $\mathcal{T}$ -invariant systems but without $\mathcal{P}$ -symmetry. Importantly, by using the Neumann's principle, we note that $\sigma_{\alpha\beta}^\gamma$ can deliver an intrinsic nonlinear Hall current in $\mathcal{T}$ -invariant systems with the crystalline point groups of 1, 2, m , 222, $mm2$, 4, -4 , 422, $4mm$, $-42m$, 3, 32, $3m$, 6, -6 , 622, $6mm$, $-6m2$, 23, 432, $-43m$. Furthermore, this intrinsic nonlinear displacement current can coexist with the $\mathcal{T}$ -even Berry-curvature-dipole driven extrinsic nonlinear Hall current [6] particularly under an AC electric field.

B. The extrinsic nonlinear displacement current

In addition to the intrinsic nonlinear displacement current, the AC semiclassical theory also delivers an extrinsic nonlinear displacement current, which is given by

$$J_\lambda^{(2;\bar{1})} \equiv \sum_n \int_k f_n^{(1)} \partial_t a_n^{(1;\lambda)} = -\omega \tau \bar{\sigma}_{\alpha\beta}^\gamma E_\alpha E_\beta^\omega, \quad (114)$$

where we have taken $\omega_1 = 0$ in Eq. (40) and summed over $\omega_2 = \pm\omega$ appeared in $a_n^{(1;\lambda)}$. By doing this, we find

$$\bar{\sigma}_{\alpha\beta}^\gamma = \sum_{nm} \int_k \partial_\alpha f_n \frac{g_{nm}^{\beta\lambda}}{\epsilon_{nm}} \quad (115)$$

by further assuming $\hbar\omega \ll \epsilon_{mn}$.

Interestingly, we find that Eq. (115) is also solely driven by the quantum metric dipole like the intrinsic nonlinear Ohmic and Hall currents. As a result, the corresponding conductivity tensors exactly respect the same symmetry transformation under the magnetic point group operations. However, we note that the nonlinear extrinsic displacement current scales with a dimensionless constant $\omega\tau$, which means that the extrinsic nonlinear current will be significantly enhanced when $\omega\tau \gg 1$ under an AC electric field while the intrinsic nonlinear Ohmic and Hall currents will not be affected.

VII. DISCUSSION AND SUMMARY

First of all, although we stop at the second order of electric field, we believe that the equivalence between both formulations can be extended to higher-order, such as the third-order. Second, we wish to mention that the response theory used in this work is formulated at length gauge. Recently, some second-order nonlinear currents [63, 92] based on the response theory particularly at velocity gauge are proposed, which can also not be reproduced by the AC semiclassical theory and are treated as the quantum correction beyond the semiclassical theory. However, based on the equivalence between both formulations, we believe that these quantum correction may also be rooted in different ways of introducing phenomenological relaxation. In addition, we wish to remark that the τ^{-1} -dependent nonlinear current obtained with response theory at the length gauge may also be related to the different scheme of phenomenological relaxation [64]. Finally, we did not discuss the consequences of the side jump and skew scattering [28, 93–96] which is beyond the scope of this work. In addition, the equivalence between both formulations with the consideration of magnetic field [97] particularly at nonlinear regimes may be explored in the future. And a detailed study about the intrinsic and extrinsic nonlinear displacement currents also will be investigated in the future.

In summary, by extending the linear AC semiclassical theory under a uniform electric field to the nonlinear regime, we show that up to the second order of the electric field, the AC semiclassical theory is equivalent to the response theory in the clean limit and this equivalence can be expected even in the DC limit when the zero-frequency divergence is properly regulated by introducing the relaxation on the same footing. In particular, in the linear order of the electric field, we show that both formulations can give the linear Drude current, intrinsic anomalous Hall current, and the intrinsic displacement current, in which the last one arising from the time variation of the first-order *positional shift* can only be driven by an AC field and is usually overlooked even in the AC transport. Furthermore, in the second order of the electric field, we show that the familiar transport effects, including the nonlinear Drude current, the non-

linear Hall currents that driven by the Berry curvature dipole and quantum metric dipole, respectively, can be obtained by both formulations. Furthermore, we show that both formulations can give the extrinsic and intrinsic nonlinear displacement currents, both of which can only appear in AC transport and are not reported yet. Notably, we find that the extrinsic nonlinear displacement current is also solely driven by the quantum metric dipole but scales with a dimensionless constant $\omega\tau$. As a result, both the quantum-metric-dipole driven intrinsic nonlinear Hall and Ohmic currents under an AC electric field can be significantly enhanced by this extrinsic nonlinear displacement current when $\omega\tau \gg 1$. Besides, we note that the $\mathcal{T}$ -even intrinsic nonlinear displacement current can coexist with the Berry-curvature-dipole driven extrinsic nonlinear Hall current under an AC electric field.

On top of this equivalence, (i) we conclude that the semiclassical theory does not ignore interband coherent effects compared to the response theory; (ii) we show that the intrinsic nonlinear Ohmic current and the non-reciprocal magnetoresistance represent the same physics, both of which arise from the second-order semiclassical wavepacket energy correction; (iii) we suggest that the energy used in the equilibrium Fermi distribution in the semiclassical theory is the unperturbed band energy by assuming that both formulations give the same intrinsic responses; (iv) we argue that the scheme of introducing relaxation in the response theory by modifying the Liouville equation for the nonequilibrium density matrix needs to be reconsidered. Our work clarifies the conceptual puzzles encountered in the emergent field of *nonlinear Hall effects* and highlights that the displacement nonlinear current responses unique to the AC transport can be important especially when a high-frequency AC driving field is applied.

VIII. APPENDIX

Appendix A: Calculation details in Section II

1. Eqs.(27-29)

With the normalized second-order wavepacket Eq.(26), the position center $\mathbf{r}_c$ can be expressed as

$$\mathbf{r}_c \equiv \langle \bar{W} | \mathbf{r} | \bar{W} \rangle = (1 + 2\delta) \langle W_0 | \mathbf{r} | W_0 \rangle + 2\text{Re} \langle W_0 | \mathbf{r} | W_1 \rangle + \langle W_1 | \mathbf{r} | W_1 \rangle + 2\text{Re} \langle W_0 | \mathbf{r} | W_2 \rangle + \mathcal{O}(E_\alpha^3). \quad (\text{A1})$$

For $\langle W_0 | \mathbf{r} | W_0 \rangle$, we find [4]

$$\langle W_0 | \mathbf{r} | W_0 \rangle = \int_{\mathbf{p}} \int_{\mathbf{p}'} C_n(\mathbf{p}) C_n^*(\mathbf{p}') e^{i(\mathbf{p}-\mathbf{p}') \cdot \mathbf{r}} \langle u'_n | \mathbf{r} | u_n \rangle = \int_{\mathbf{p}} \int_{\mathbf{p}'} C_n(\mathbf{p}) C_n^*(\mathbf{p}') [-i\partial_{\mathbf{p}} + \mathcal{A}_n] \delta(\mathbf{p} - \mathbf{p}') \partial_{\mathbf{p}_c} \gamma(\mathbf{p}_c) + \mathcal{A}_n(\mathbf{p}_c). \quad (\text{A2})$$

where we have used $|C_n(\mathbf{p})|^2 = \delta(\mathbf{p} - \mathbf{p}_c)$. In addition, we have assumed that $C_n(\mathbf{p}) = |C_n(\mathbf{p})| e^{-i\gamma(\mathbf{p})}$ and used that

$$\begin{aligned} \int_{\mathbf{p}} \int_{\mathbf{p}'} C_n(\mathbf{p}) C_n^*(\mathbf{p}') (-i\partial_{\mathbf{p}}) \delta(\mathbf{p} - \mathbf{p}') &= \int_{\mathbf{p}} [i\partial_{\mathbf{p}} C_n(\mathbf{p})] C_n^*(\mathbf{p}) = \int_{\mathbf{p}} \partial_{\mathbf{p}} \gamma(\mathbf{p}) |C_n(\mathbf{p})|^2 + \int_{\mathbf{p}} |C_n(\mathbf{p})| i\partial_{\mathbf{p}} |C_n(\mathbf{p})| \\ &= \partial_{\mathbf{p}_c} \gamma(\mathbf{p}_c) + \frac{i}{2} \partial_{\mathbf{p}} \left(\int_{\mathbf{p}} |C_n(\mathbf{p})|^2 \right) = \partial_{\mathbf{p}_c} \gamma(\mathbf{p}_c). \end{aligned} \quad (\text{A3})$$

For $2\text{Re} \langle W_0 | \mathbf{r} | W_1 \rangle$, we find

$$2\text{Re} \langle W_0 | \mathbf{r} | W_1 \rangle = 2\text{Re} \left[\int_{\mathbf{p}} \int_{\mathbf{p}'} \sum_{m \neq n} M_{mn}^{(1)}(\mathbf{p}) C_n(\mathbf{p}) C_n^*(\mathbf{p}') e^{i(\mathbf{p}-\mathbf{p}') \cdot \mathbf{r}} \langle u'_n | \mathbf{r} | u_m \rangle \right] = 2\text{Re} \sum_m M_{mn}^{(1)}(\mathbf{p}_c) \mathbf{r}_{nm}(\mathbf{p}_c). \quad (\text{A4})$$

Similarly, for $2\text{Re} \langle W_0 | \mathbf{r} | W_2 \rangle$, we find

$$2\text{Re} \langle W_0 | \mathbf{r} | W_2 \rangle = 2\text{Re} \left[\int_{\mathbf{p}} \int_{\mathbf{p}'} \sum_{m \neq n} M_{mn}^{(2)}(\mathbf{p}) C_n(\mathbf{p}) C_n^*(\mathbf{p}') e^{i(\mathbf{p}-\mathbf{p}') \cdot \mathbf{r}} \langle u'_n | \mathbf{r} | u_m \rangle \right] = 2\text{Re} \sum_m M_{mn}^{(2)}(\mathbf{p}_c) \mathbf{r}_{nm}(\mathbf{p}_c). \quad (\text{A5})$$

Finally, for $\langle W_1 | \mathbf{r} | W_1 \rangle$, we find:

$$\begin{aligned} \langle W_1 | \mathbf{r} | W_1 \rangle &= \int_{\mathbf{p}} \int_{\mathbf{p}'} \sum_{m \neq n} \sum_{l \neq n} M_{mn}^{(1)}(\mathbf{p}) M_{ln}^{(1)*}(\mathbf{p}') C_n(\mathbf{p}) C_n^*(\mathbf{p}') e^{i(\mathbf{p}-\mathbf{p}') \cdot \mathbf{r}} \langle u'_l | \mathbf{r} | u_m \rangle \\ &= \int_{\mathbf{p}} \int_{\mathbf{p}'} \sum_{m \neq n} \sum_{l \neq n} M_{mn}^{(1)}(\mathbf{p}) M_{ln}^{(1)*}(\mathbf{p}') C_n(\mathbf{p}) C_n^*(\mathbf{p}') [-i\delta_{ln} \partial_{\mathbf{p}} + \mathcal{A}_{lm}] \delta(\mathbf{p}' - \mathbf{p}) \\ &= \int_{\mathbf{p}} \sum_{m \neq n} M_{mn}^{(1)*}(\mathbf{p}) C_n^*(\mathbf{p}) i\partial_{\mathbf{p}} \left[M_{mn}^{(1)}(\mathbf{p}) C_n(\mathbf{p}) \right] + \sum_{l \neq n} \sum_{m \neq n} M_{ln}^{(1)*}(\mathbf{p}_c) \mathcal{A}_{lm}(\mathbf{p}_c) M_{mn}^{(1)}(\mathbf{p}_c) \\ &= \sum_{m \neq n} M_{mn}^{(1)*}(\mathbf{p}_c) i\partial_{\mathbf{p}_c} M_{mn}^{(1)}(\mathbf{p}_c) + \partial_{\mathbf{p}_c} \gamma(\mathbf{p}_c) \sum_{m \neq n} M_{mn}^{(1)*}(\mathbf{p}_c) M_{mn}^{(1)}(\mathbf{p}_c) + \sum_{l \neq n} \sum_{m \neq n} M_{ln}^{(1)*}(\mathbf{p}_c) \mathcal{A}_{lm}(\mathbf{p}_c) M_{mn}^{(1)}(\mathbf{p}_c) \\ &\quad + \sum_{m \neq n} \int_{\mathbf{p}} M_{mn}^{(1)*}(\mathbf{p}) M_{mn}^{(1)}(\mathbf{p}) \frac{i}{2} \partial_{\mathbf{p}} |C_n(\mathbf{p})|^2 \\ &= \sum_{m \neq n} M_{mn}^{(1)*}(\mathbf{p}_c) i\partial_{\mathbf{p}_c} M_{mn}^{(1)}(\mathbf{p}_c) + \partial_{\mathbf{p}_c} \gamma(\mathbf{p}_c) \sum_{m \neq n} M_{mn}^{(1)*}(\mathbf{p}_c) M_{mn}^{(1)}(\mathbf{p}_c) + \sum_{l \neq n} \sum_{m \neq n} M_{ln}^{(1)*}(\mathbf{p}_c) \mathcal{A}_{lm}(\mathbf{p}_c) M_{mn}^{(1)}(\mathbf{p}_c) \\ &\quad - \sum_{m \neq n} \frac{1}{2} i\partial_{\mathbf{p}_c} \left[M_{mn}^{(1)*}(\mathbf{p}_c) M_{mn}^{(1)}(\mathbf{p}_c) \right] \\ &= \text{Re} \sum_{m \neq n} M_{mn}^{(1)*}(\mathbf{p}_c) i\partial_{\mathbf{p}_c} M_{mn}^{(1)}(\mathbf{p}_c) + \partial_{\mathbf{p}_c} \gamma(\mathbf{p}_c) \sum_{m \neq n} M_{mn}^{(1)*}(\mathbf{p}_c) M_{mn}^{(1)}(\mathbf{p}_c) + \sum_{l \neq n} \sum_{m \neq n} M_{ln}^{(1)*}(\mathbf{p}_c) \mathcal{A}_{lm}(\mathbf{p}_c) M_{mn}^{(1)}(\mathbf{p}_c). \end{aligned} \quad (\text{A6})$$

Collecting these results, we obtain (for simplicity, we will drop the explicit dependence on $\mathbf{p}_c$ below)

$$\mathbf{r}_c = \partial_{\mathbf{p}_c} \gamma + \mathcal{A}_n + \mathbf{a}_n^{(1)} + \mathbf{a}_n^{(2)}, \quad (\text{A7})$$

as given by Eq.(27) in the main text. Here $\mathbf{a}_n^{(i)}$ is the i th *positional shift* given by

$$\mathbf{a}_n^{(1)} \equiv 2\text{Re}\langle W_0 | \mathbf{r} | W_1 \rangle = 2\text{Re} \sum_m M_{mn}^{(1)} \mathbf{r}_{nm} \quad (\text{A8})$$

and

$$\begin{aligned} \mathbf{a}_n^{(2)} &\equiv 2\delta\langle W_0 | \mathbf{r} | W_0 \rangle + 2\text{Re}\langle W_0 | \mathbf{r} | W_2 \rangle + \langle W_1 | \mathbf{r} | W_1 \rangle \\ &= - \sum_{m \neq n} M_{mn}^{(1)} \mathcal{A}_n M_{mn}^{(1)*} + 2\text{Re} \sum_{m \neq n} M_{mn}^{(2)} \mathbf{r}_{nm} + \text{Re} \sum_{m \neq n} M_{mn}^{(1)*} i \partial_{\mathbf{p}_c} M_{mn}^{(1)} + \sum_{l \neq n} \sum_{m \neq n} M_{ln}^{(1)*} \mathcal{A}_m M_{mn}^{(1)} \\ &= \text{Re} \sum_m M_{mn}^{(1)*} i \mathcal{D}_{mn}^{\mathbf{p}_c} M_{mn}^{(1)} + \sum_{ml} M_{ln}^{(1)*} \mathbf{r}_{lm} M_{mn}^{(1)} + 2\text{Re} \sum_m M_{mn}^{(2)} \mathbf{r}_{nm} \\ &= -\text{Re} \sum_m M_{mn}^{(1)} i \mathcal{D}_{nm}^{\mathbf{p}_c} M_{mn}^{(1)*} + \sum_{ml} M_{ln}^{(1)*} \mathbf{r}_{lm} M_{mn}^{(1)} + 2\text{Re} \sum_m M_{mn}^{(2)} \mathbf{r}_{nm} \end{aligned} \quad (\text{A9})$$

where $2\delta\partial_{\mathbf{p}_c} \gamma$ is exactly canceled out with the second term of Eq.(A6). Furthermore, using the Levi-Civita symbol and Eq.(20), we finally find:

$$\mathbf{a}_n^{(1;\lambda)} = 2\text{Re} \sum_m \sum_{\omega_1} \frac{r_{nm}^\lambda r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} E_\alpha^{\omega_1}, \quad (\text{A10})$$

where $E_\alpha^{\omega_1} \equiv e^{-i(\omega_1 + i\xi)t}$, as given by Eq.(28) in the main text. Furthermore, by using Eq.(20) and Eq.(22), we find

$$\begin{aligned} \mathbf{a}_n^{(2;\lambda)} &= \text{Re} \sum_{\omega_1 \omega_2} \sum_{ml} \frac{2r_{nm}^\lambda r_{ml}^\beta r_{ln}^\alpha}{(\omega_1 - \epsilon_{ln} + i\xi)(\omega_\Sigma - \epsilon_{mn} + 2i\xi)} E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ &\quad + \sum_{\omega_1 \omega_2} \sum_{ml} \frac{-r_{nl}^\beta r_{lm}^\lambda r_{mn}^\alpha}{(\omega_2 - \epsilon_{nl} + i\xi)(\omega_1 - \epsilon_{mn} + i\xi)} E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ &\quad + \text{Re} \sum_{\omega_1 \omega_2} \sum_m \frac{2r_{nm}^\lambda}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} i \mathcal{D}_{mn}^\beta \left(\frac{r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} \right) E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ &\quad + \text{Re} \sum_{\omega_1 \omega_2} \sum_m \frac{r_{mn}^\beta}{\omega_2 - \epsilon_{mn} + i\xi} i \mathcal{D}_{nm}^\lambda \left(\frac{r_{nm}^\alpha}{\omega_1 - \epsilon_{nm} + i\xi} \right) E_\alpha^{\omega_1} E_\beta^{\omega_2} \end{aligned} \quad (\text{A11})$$

as given by Eq.(29) in the main text, where we have used that

$$M_{mn}^{(1)*} = \sum_{\omega_1} \frac{r_{nm}^\alpha E_\alpha e^{i(\omega_1 - i\xi)t}}{\omega_1 - \epsilon_{mn} - i\xi} = \sum_{\omega_1} \frac{r_{nm}^\alpha E_\alpha e^{-i(\omega_1 + i\xi)t}}{-\omega_1 - \epsilon_{mn} - i\xi} = -M_{nm}^{(1)} \quad (\text{A12})$$

due to $\sum_{\omega_1 = \pm\omega} f(\omega_1) = \sum_{\omega_1 = \pm\omega} f(-\omega_1)$ for an arbitrary function $f(\omega_1)$.

2. Eqs.(36-37)

With the second-order wavepacket Eq.(26), the energy for the wavepacket can be expressed as

$$\begin{aligned} \bar{\epsilon}_n &\equiv \langle W_0 | i\partial_t | W_0 \rangle - \langle \bar{W} | (i\partial_t - H) | \bar{W} \rangle \\ &= [-\langle W_0 | i\partial_t | \delta | W_0 \rangle] - \delta \langle W_0 | i\partial_t | W_0 \rangle - \langle W_1 | i\partial_t | W_1 \rangle \\ &\quad + [(1 + 2\delta) \langle W_0 | H_0 | W_0 \rangle + \langle W_1 | H_0 | W_1 \rangle + \mathbf{E} \cdot \langle \bar{W} | \mathbf{r} | \bar{W} \rangle] + \mathcal{O}(E^3) \\ &= [-i\partial_t \delta - 2\delta\partial_t \gamma - \langle W_1 | i\partial_t | W_1 \rangle] + \left[(1 + 2\delta)\epsilon_n + \sum_m M_{mn}^{(1)} M_{mn}^{(1)*} \epsilon_m + \mathbf{E} \cdot \mathbf{r}_c \right] + \mathcal{O}(E^3), \end{aligned} \quad (\text{A13})$$

where we have used Eq.(31), $\langle W_0|W_1\rangle = 0$, and $\langle W_0|W_2\rangle = 0$. Note that

$$\begin{aligned}
\langle W_1|i\partial_t|W_1\rangle &= \int_{\mathbf{p}} \int_{\mathbf{p}'} \sum_{m \neq n} \sum_{l \neq n} e^{i(\mathbf{p}-\mathbf{p}') \cdot \mathbf{r}} M_{ln}^{(1)*}(\mathbf{p}') \langle u_l' | C_n(\mathbf{p}') i\partial_t \left(M_{mn}^{(1)}(\mathbf{p}) C_n(\mathbf{p}) | u_m \rangle \right) \\
&= \sum_m M_{mn}^{(1)} i\partial_t M_{mn}^{(1)*} + \int_{\mathbf{p}} \sum_m M_{mn}^{(1)*}(\mathbf{p}) M_{mn}^{(1)}(\mathbf{p}) C_n(\mathbf{p}) i\partial_t C_n(\mathbf{p}) \\
&= \sum_m M_{mn}^{(1)} i\partial_t M_{mn}^{(1)*} + \partial_t \gamma \sum_m M_{mn}^{(1)} M_{mn}^{(1)*} + \int_{\mathbf{p}} \sum_m M_{mn}^{(1)*}(\mathbf{p}) M_{mn}^{(1)}(\mathbf{p}) \frac{i}{2} \partial_t |C_n(\mathbf{p})|^2 \\
&= \sum_m M_{mn}^{(1)} i\partial_t M_{mn}^{(1)*} + \partial_t \gamma \sum_m M_{mn}^{(1)} M_{mn}^{(1)*} + \sum_m \frac{i}{2} \partial_t \left(\int_{\mathbf{p}} M_{mn}^{(1)*}(\mathbf{p}) M_{mn}^{(1)}(\mathbf{p}) |C_n(\mathbf{p})|^2 \right) \\
&\quad - \sum_m \int_{\mathbf{p}} \frac{i}{2} \partial_t \left(M_{mn}^{(1)*}(\mathbf{p}) M_{mn}^{(1)}(\mathbf{p}) \right) |C_n(\mathbf{p})|^2 \\
&= \sum_m M_{mn}^{(1)} i\partial_t M_{mn}^{(1)*} - 2\delta \partial_t \gamma,
\end{aligned} \tag{A14}$$

where we have used Eq.(25). Substituting Eq.(A14) into Eq.(A15), we find:

$$\begin{aligned}
\bar{\epsilon}_n &= \epsilon_n + \mathbf{E} \cdot \mathbf{r}_c + \frac{1}{2} i\partial_t \left(\sum_m M_{mn}^{(1)} M_{mn}^{(1)*} \right) - \sum_m M_{mn}^{(1)} i\partial_t M_{mn}^{(1)*} + \sum_m \epsilon_{mn} M_{mn}^{(1)} M_{mn}^{(1)*} + \mathcal{O}(E^3) \\
&= \epsilon_n + \mathbf{E} \cdot \mathbf{r}_c + \frac{1}{2} \sum_m \left[\left(i\partial_t M_{mn}^{(1)} \right) M_{mn}^{(1)*} - M_{mn}^{(1)} i\partial_t M_{mn}^{(1)*} \right] + \sum_m \epsilon_{mn} M_{mn}^{(1)} M_{mn}^{(1)*} + \mathcal{O}(E^3) \\
&= \epsilon_n + \mathbf{E} \cdot \mathbf{r}_c + \text{Re} \sum_m \left[\left(i\partial_t M_{mn}^{(1)} \right) M_{mn}^{(1)*} + \epsilon_{mn} M_{mn}^{(1)} M_{mn}^{(1)*} \right] \\
&= \epsilon_n + \mathbf{E} \cdot \mathbf{r}_c + \epsilon_n^{(2)} + \mathcal{O}(E^3),
\end{aligned} \tag{A15}$$

as given by Eq.(36) in the main text, where $\mathbf{r}_c \equiv \langle \bar{W} | \mathbf{r} | \bar{W} \rangle$ and

$$\begin{aligned}
\epsilon_n^{(2)} &\equiv \text{Re} \sum_m \left[\left(i\partial_t M_{mn}^{(1)} \right) M_{mn}^{(1)*} + \epsilon_{mn} M_{mn}^{(1)} M_{mn}^{(1)*} \right] = -\text{Re} \sum_m \sum_{\omega_1 \omega_2} \frac{(\omega_1 + i\xi + \epsilon_{mn}) r_{mn}^\alpha r_{nm}^\beta E_\alpha^{\omega_1} E_\beta^{\omega_2}}{(\omega_1 - \epsilon_{mn} + i\xi)(\omega_2 - \epsilon_{nm} + i\xi)} \\
&= -\text{Re} \sum_m \sum_{\omega_1 \omega_2} \frac{r_{mn}^\alpha r_{nm}^\beta E_\alpha^{\omega_1} E_\beta^{\omega_2}}{\omega_2 - \epsilon_{nm} + i\xi} = -\frac{1}{2} \sum_{\omega_1} a_n^{(1;\alpha)} E_\alpha^{\omega_1},
\end{aligned} \tag{A16}$$

as given by Eq.(37) in the main text, where we have used $M_{mn}^{(1)*} = -M_{nm}^{(1)}$ and Eq.(28). Note that only the first term of Eq.(A15) contains $\mathbf{r}_c$ while Eq.(A16) depends only on $\mathbf{k}_c$.

Appendix B: The solution for Boltzmann equation

In this appendix, we will derive Eq.(40) and Eq.(41) given in the main text, which are obtained by solving the Boltzmann equation in presence of the electric field. For completeness, the Boltzmann equation is reproduced as follows:

$$\partial_t \bar{f}_n + \eta(\bar{f}_n - f_n^{(0)}) = E_\alpha^{\omega_1} \partial_\alpha \bar{f}_n, \tag{B1}$$

where the relaxation time approximation $\tau = 1/\eta$ is adopted. Eq.(B1) can be iteratively solved from the equilibrium distribution function $f_n^{(0)} = f_n = 1/[e^{(\epsilon_n - \mu)/k_B T} + 1]$ without the electric field, where μ is the chemical potential, k_B is the Boltzmann constant, and T is the temperature. Straightforwardly, by inserting $\bar{f}_n = f_n^{(0)} + f_n^{(1)} + f_n^{(2)} + \dots$ into Eq.(B1), where $f_n^{(i)} \propto E_\alpha^{(i)}$, we obtain the following recursive first-order inhomogeneous linear differential equation equations for the nonequilibrium distribution:

$$\partial_t f_n^{(1)} + \eta f_n^{(1)} = E_\alpha^{\omega_1} \partial_\alpha f_n^{(0)}, \tag{B2}$$

$$\partial_t f_n^{(2)} + \eta f_n^{(2)} = E_\beta^{\omega_2} \partial_\beta f_n^{(1)}, \tag{B3}$$

which are truncated at the second order of the electric field. Note that $f_n^{(i)} = 0$ with $i = 1, 2$ in the absence of the electric field ($t \rightarrow -\infty$) is the boundary condition for Eqs.(B2-B3). Recall that for the first-order inhomogeneous linear differential equation $\partial_t y(t) + p(t)y(t) = q(t)$, the general solution is given by

$$y(t) = e^{-\int p(t)dt} \left(C + \int q(t) e^{\int p(t)dt} dt \right). \quad (\text{B4})$$

Using Eq.(B4), for Eq.(B2), we immediately obtain

$$f_n^{(1)} = e^{-\eta t} \left(C + E_\alpha \partial_\alpha f_n^{(0)} \int e^{-i[\omega_1 + i(\eta + \xi)]t} dt \right) = \frac{E_\alpha^{\omega_1} \partial_\alpha f_n^{(0)}}{-i[\omega_1 + i(\eta + \xi)]} = \frac{i \partial_\alpha f_n^{(0)} E_\alpha^{\omega_1}}{\omega_1 + i(\eta + \xi)}, \quad (\text{B5})$$

where $p(t) = \eta$ and $q(t) = E_\alpha^{\omega_1} \partial_\alpha f_n^{(0)}$ in Eq.(B4) and $C = 0$ fixed by the boundary condition. Similarly, for Eq.(B3), we obtain

$$\begin{aligned} f_n^{(2)} &= e^{-\eta t} \left(C + \frac{i \partial_{\beta\alpha}^2 f_n^{(0)}}{\omega_1 + i(\eta + \xi)} \int e^{-i[\omega_\Sigma + i(\eta + 2\xi)]t} dt \right) = \frac{E_\alpha^{\omega_1} E_\beta^{\omega_2} i \partial_{\alpha\beta}^2 f_n^{(0)}}{[\omega_1 + i(\eta + \xi)] [-i\omega_\Sigma + (\eta + 2\xi)]} \\ &= \frac{-\partial_{\alpha\beta}^2 f_n^{(0)} E_\alpha^{\omega_1} E_\beta^{\omega_2}}{[\omega_1 + i(\eta + \xi)] [\omega_\Sigma + i(\eta + 2\xi)]}, \end{aligned} \quad (\text{B6})$$

where $q(t) = f_n^{(1)}$ has been used in Eq.(B4). Eq.(B5) and Eq.(41) are given by Eq.(40) and Eq.(41) in the main text, respectively.

Appendix C: Response theory

In this appendix, we first derive the expressions for the density matrix element by iteratively solving the quantum Liouville equation. With the density matrix element, we further derive the linear and nonlinear conductivities given in the main text.

1. The density matrix element

The quantum Liouville equation for the density matrix in Bloch basis is reproduced as

$$i \partial_t \rho_{mn} = [H, \rho]_{mn} = \epsilon_{mn} \rho_{mn} + [r^\alpha, \rho]_{mn} E_\alpha^{\omega_1}, \quad (\text{C1})$$

where $\epsilon_{mn} = \epsilon_m - \epsilon_n$ and

$$[r^\alpha, \rho]_{mn} = i \mathcal{D}_{mn}^\alpha \rho_{mn} + \sum_l (r_{ml}^\alpha \rho_{ln} - \rho_{ml} r_{ln}^\alpha). \quad (\text{C2})$$

Here $\mathcal{D}_{mn}^\alpha \equiv \partial_\alpha - i(\mathcal{A}_m^\alpha - \mathcal{A}_n^\alpha)$ is the $U(1)$ covariant derivative. Eq.(C1) can also be solved iteratively. Particularly, by writing $\rho_{mn} = \sum_i \rho_{mn}^{(i)}$, where $\rho_{mn}^{(i)} \propto E_\alpha^{(i)}$ and $\rho_{mn}^{(0)} = \delta_{mn} f_n$, we obtain the following recursive first-order inhomogeneous linear differential equations up to the second order of the electric field:

$$\partial_t \rho_{mn}^{(1)} + i \epsilon_{mn} \rho_{mn}^{(1)} = -i [r^\alpha, \rho^{(0)}]_{mn} E_\alpha^{\omega_1} \quad (\text{C3})$$

$$\partial_t \rho_{mn}^{(2)} + i \epsilon_{mn} \rho_{mn}^{(2)} = -i [r^\beta, \rho^{(1)}]_{mn} E_\beta^{\omega_2}, \quad (\text{C4})$$

where the boundary condition is $\rho_{mn}^{(i)}(t) = 0$ with $i \geq 1$ when $t \rightarrow -\infty$, which implies that the AC electric field is applied adiabatically.

For Eq.(C3), by taking $p(t) = i \epsilon_{mn}$ and

$$q(t) = -i [r^\alpha, \rho^{(0)}]_{mn} E_\alpha^{\omega_1} = \left[\mathcal{D}_{mn}^\alpha \rho_{mn}^{(0)} - i \sum_l (r_{ml}^\alpha \rho_{ln}^{(0)} - \rho_{ml}^{(0)} r_{ln}^\alpha) \right] E_\alpha^{\omega_1} = (\delta_{mn} \partial_\alpha f_m - i f_{nm} r_{mn}^\alpha) E_\alpha^{\omega_1} \quad (\text{C5})$$

in Eq.(B4), where $f_{nm} = f_n - f_m$, we find

$$\begin{aligned}\rho_{mn}^{(1)} &= e^{-i\epsilon_{mn}t} \left[C + (\delta_{mn}\partial_\alpha f_m - i f_{nm} r_{mn}^\alpha) E_\alpha \int e^{-i(\omega_1 - \epsilon_{mn} + i\xi)t} dt \right] = \frac{\delta_{mn}\partial_\alpha f_m - i f_{nm} r_{mn}^\alpha}{-i(\omega_1 - \epsilon_{mn} + i\xi)} E_\alpha^{\omega_1} \\ &= \frac{i\delta_{mn}\partial_\alpha f_m E_\alpha^{\omega_1}}{\omega_1 + i\xi} + \frac{f_{nm} r_{mn}^\alpha E_\alpha^{\omega_1}}{\omega_1 - \epsilon_{mn} + i\xi},\end{aligned}\quad (C6)$$

Note that $C = 0$ is fixed by the boundary condition. Similarly, for Eq.(C4), we note that $q(t) = i\epsilon_{mn}$ and

$$\begin{aligned}q(t) &= -i[r^\beta, \rho^{(1)}]_{mn} E_\beta^{\omega_2} = \left[\mathcal{D}_{mn}^\beta \rho_{mn}^{(1)} - i \sum_l \left(r_{ml}^\beta \rho_{ln}^{(1)} - \rho_{ml}^{(1)} r_{ln}^\beta \right) \right] E_\beta^{\omega_2} \\ &= \left[\frac{i\delta_{mn}\partial_{\alpha\beta}^2 f_m}{\omega_1 + i\xi} + \mathcal{D}_{mn}^\beta \left(\frac{f_{nm} r_{mn}^\alpha}{\omega_1 - \omega_{mn} + i\xi} \right) \right] E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ &\quad - i \left[\sum_l \left(\frac{r_{ml}^\beta i\delta_{ln}\partial_\alpha f_l}{\omega_1 + i\xi} + \frac{r_{ml}^\beta f_{nl} r_{ln}^\alpha}{\omega_1 - \epsilon_{ln} + i\xi} \right) - \sum_l \left(\frac{i\delta_{ml}\partial_\alpha f_m r_{ln}^\beta}{\omega_1 + i\xi} + \frac{f_{lm} r_{ml}^\alpha r_{ln}^\beta}{\omega_1 - \epsilon_{ml} + i\xi} \right) \right] E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ &\equiv T E_\alpha^{\omega_1} E_\beta^{\omega_2},\end{aligned}\quad (C7)$$

where

$$T = \frac{i\delta_{mn}\partial_{\alpha\beta}^2 f_m}{\omega_1 + i\xi} + \mathcal{D}_{mn}^\beta \left(\frac{f_{nm} r_{mn}^\alpha}{\omega_1 - \omega_{mn} + i\xi} \right) + \frac{r_{mn}^\beta \partial_\alpha f_{nm}}{\omega_1 + i\xi} - i \sum_l \left(\frac{r_{ml}^\beta r_{ln}^\alpha f_{nl}}{\omega_1 - \epsilon_{ln} + i\xi} - \frac{f_{lm} r_{ml}^\alpha r_{ln}^\beta}{\omega_1 - \epsilon_{ml} + i\xi} \right). \quad (C8)$$

Also by inserting the explicit expressions for $p(t)$ and $q(t)$ into Eq.(B4), we find:

$$\begin{aligned}\rho_{mn}^{(2)} &= e^{-i\epsilon_{mn}t} \left(C + T E_\alpha E_\beta \int e^{-i(\omega_\Sigma - \epsilon_{mn} + 2i\xi)t} \right) = \frac{T E_\alpha^{\omega_1} E_\beta^{\omega_2}}{-i(\omega_\Sigma - \epsilon_{mn} + 2i\xi)} = \frac{i T E_\alpha^{\omega_1} E_\beta^{\omega_2}}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} \\ &= \left[\frac{-\delta_{mn}\partial_{\alpha\beta}^2 f_m}{(\omega_1 + i\xi)(\omega_\Sigma + 2i\xi)} + \frac{1}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} \mathcal{D}_{mn}^\beta \left(\frac{i f_{nm} r_{mn}^\alpha}{\omega_1 - \omega_{mn} + i\xi} \right) + \frac{i r_{mn}^\beta \partial_\alpha f_{nm}}{(\omega_1 + i\xi)(\omega_\Sigma - \epsilon_{mn} + 2i\xi)} \right] E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ &\quad + \sum_l \frac{1}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} \left(\frac{r_{ml}^\beta r_{ln}^\alpha f_{nl}}{\omega_1 - \epsilon_{ln} + i\xi} - \frac{f_{lm} r_{ml}^\alpha r_{ln}^\beta}{\omega_1 - \epsilon_{ml} + i\xi} \right) E_\alpha^{\omega_1} E_\beta^{\omega_2},\end{aligned}\quad (C9)$$

where $\omega_\Sigma = \omega_1 + \omega_2$. With Eq.(C6) and Eq.(C9), we are ready to calculate the conductivity up to the second order.

2. The conductivity

In this subsection, we will derive Eqs.(53-55) and Eqs.(57-60) discussed in the main text. In general, the current density can also be expressed as $J_\lambda(t) = \sum_{i=1}^\infty J_\lambda^{(i)}(t)$, where $J_\lambda^{(i)}(t) \propto E_\alpha^{(i)}$ particularly with

$$J_\lambda^{(i)} \equiv \text{Tr}[\rho \hat{v}^\lambda] = \sum_{mn} \int_k \rho_{mn}^{(i)} v_{nm}^\lambda. \quad (C10)$$

At the first order of the applied electric field, by substituting Eq.(C6) into Eq.(C10), we find that $J_\lambda^{(1)} = \sum_{mn} \int_k \rho_{mn}^{(1)} v_{nm}^\lambda \equiv \sigma_{\lambda\alpha}^{(1)}(\omega_1) E_\alpha^{\omega_1}$, where the conductivity $\sigma_{\lambda\alpha}^{(1)}(\omega)$ is

$$\begin{aligned}\sigma_{\lambda\alpha}^{(1)}(\omega_1) &= \sum_n \int_k \frac{i v_n^\lambda \partial_\alpha f_n}{\omega_1 + i\xi} + \sum_{mn} \int_k \frac{f_{nm} r_{mn}^\alpha v_{nm}^\lambda}{\omega_1 - \epsilon_{mn} + i\xi} = \sigma_{\lambda\alpha}^{(1;1)} + \sum_{mn} \int_k i f_{nm} r_{mn}^\alpha r_{nm}^\lambda \left(1 - \frac{\omega_1 + i\xi}{\omega_1 - \epsilon_{mn} + i\xi} \right) \\ &= \sigma_{\lambda\alpha}^{(1;1)} + \sum_n \int_k f_n \Omega_n^{\lambda\alpha} - \sum_{nm} \int_k \frac{i(\omega_1 + i\xi) f_{nm} r_{mn}^\alpha r_{nm}^\lambda}{\omega_1 - \epsilon_{mn} + i\xi} = \sigma_{\lambda\alpha}^{(1;1)} + \sigma_{\lambda\alpha}^{(1;0)} + \sigma_{\lambda\alpha}^{(1;\bar{1})},\end{aligned}\quad (C11)$$

as given by Eq.(52) in the main text. Here we have used $v_{nm}^\lambda = i\epsilon_{nm} r_{nm}^\lambda$ for $m \neq n$ and $\epsilon_{nm}/(\omega_1 - \epsilon_{mn} + i\xi) = 1 - (\omega_1 + i\xi)/(\omega_1 - \epsilon_{mn} + i\xi)$. In addition, we have defined $\Omega_n^{\lambda\alpha} = \sum_m i(r_{nm}^\lambda r_{mn}^\alpha - r_{nm}^\alpha r_{mn}^\lambda)$ and

$$\sigma_{\lambda\alpha}^{(1;1)} = \sum_n \int_k \frac{i v_n^\lambda \partial_\alpha f_n}{\omega_1 + i\xi}, \quad \sigma_{\lambda\alpha}^{(1;0)} = \sum_n \int_k f_n \Omega_n^{\lambda\alpha}, \quad \sigma_{\lambda\alpha}^{(1;\bar{1})} = \sum_{nm} \int_k \frac{-i(\omega_1 + i\xi) f_{nm} r_{mn}^\alpha r_{nm}^\lambda}{\omega_1 - \epsilon_{mn} + i\xi}, \quad (C12)$$

as given by Eqs.(53-55) in the main text.

Similarly, at the second order of the electric field, by substituting Eq.(C9) into Eq.(C10), we have $J_\lambda^{(2)} = \sum_{mn} \int_k \rho_{mn}^{(2)} v_{nm}^\lambda \equiv \sigma_{\lambda\alpha\beta}^{(2)}(\omega_1, \omega_2) E_\alpha^{\omega_1} E_\beta^{\omega_2}$, where $\sigma_{\lambda\alpha\beta}^{(2)}(\omega_1, \omega_2) \equiv \sigma_{\lambda\alpha\beta}^{(2,ee)} + \sigma_{\lambda\alpha\beta}^{(2,ei)} + \sigma_{\lambda\alpha\beta}^{(2,ie)} + \sigma_{\lambda\alpha\beta}^{(2,ii)}$ with

$$\sigma_{\lambda\alpha\beta}^{(2,ee)} = \sum_{mnl} \int_k \mathcal{V}_{nm}^\lambda \left(\frac{r_{ml}^\beta r_{ln}^\alpha f_{nl}}{\omega_1 - \epsilon_{ln} + i\xi} - \frac{f_{lm} r_{ml}^\alpha r_{ln}^\beta}{\omega_1 - \epsilon_{ml} + i\xi} \right), \quad (C13)$$

$$\sigma_{\lambda\alpha\beta}^{(2,ei)} = \sum_{mn} \int_k \mathcal{V}_{nm}^\lambda \left(\frac{ir_{mn}^\beta \partial_\alpha f_{nm}}{\omega_1 + i\xi} \right), \quad (C14)$$

$$\sigma_{\lambda\alpha\beta}^{(2,ie)} = \sum_{mn} \int_k \mathcal{V}_{nm}^\lambda \mathcal{D}_{mn}^\beta \left(\frac{if_{nm} r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} \right), \quad (C15)$$

$$\sigma_{\lambda\alpha\beta}^{(2,ii)} = \sum_n \int_k \frac{-v_n^\lambda \partial_{\alpha\beta}^2 f_n}{(\omega_1 + i\xi)(\omega_\Sigma + 2i\xi)}, \quad (C16)$$

as given by Eqs.(57-60) in the main text. Here $\mathcal{V}_{nm}^\lambda \equiv v_{nm}^\lambda / (\omega_\Sigma - \epsilon_{mn} + 2i\xi)$.

3. The phenomenological inclusion of relaxation

By phenomenologically including the relaxation into the quantum Liouville equation Eq. (4), we find:

$$\partial_t \rho_{mn}^{(1)} + (i\epsilon_{mn} + \eta) \rho_{mn}^{(1)} = \left[\mathcal{D}_\alpha^{mn} \rho_{mn}^{(0)} - i \sum_l \left(r_{ml}^\alpha \rho_{ln}^{(0)} - \rho_{ml}^{(0)} r_{ln}^\alpha \right) \right] E_\alpha^{\omega_1}, \quad (C17)$$

$$\partial_t \rho_{mn}^{(2)} + (i\epsilon_{mn} + \eta) \rho_{mn}^{(2)} = \left[\mathcal{D}_\beta^{mn} \rho_{mn}^{(1)} - i \sum_l \left(r_{ml}^\beta \rho_{ln}^{(1)} - \rho_{ml}^{(1)} r_{ln}^\beta \right) \right] E_\beta^{\omega_2}, \quad (C18)$$

where $\eta = 1/\tau$. As a result, by solving Eq.(C17), we find

$$\rho_{mn}^{(1)}(\omega_1) = \frac{\delta_{mn} \partial_\alpha f_m - if_{nm} r_{mn}^\alpha}{-i[\omega_1 - \epsilon_{mn} + i(\xi + \eta)]} E_\alpha^{\omega_1}. \quad (C19)$$

In the DC limit, this equation becomes

$$\rho_{mn}^{(1)} = \tau \delta_{mn} \partial_\alpha f_m E_\alpha + \frac{f_{nm} r_{mn}^\alpha}{-\epsilon_{mn} + i\eta} E_\alpha, \quad (C20)$$

where the second term gives the unphysical linear conductivity

$$\sigma_{\lambda\alpha}^H = \sum_{mn} \int_k \frac{f_{nm} r_{mn}^\alpha v_{nm}^\lambda}{-\epsilon_{mn} + i\eta} = 2 \sum_{mn} \int_k f_n \text{Re} \frac{r_{mn}^\alpha v_{nm}^\lambda}{-\epsilon_{mn} + i/\tau}, \quad (C21)$$

as given by Eq.(105) in the main text. However, if we further assume $\eta \rightarrow \bar{\eta} = \delta_{mn}/\tau$ and then we have

$$\rho_{mn}^{(1)}(\omega_1) = \frac{i\delta_{mn} \partial_\alpha f_m E_\alpha^{\omega_1}}{\omega_1 + i(\eta + \xi)} + \frac{f_{nm} r_{mn}^\alpha E_\alpha^{\omega_1}}{\omega_1 - \epsilon_{mn} + i\xi} \quad (C22)$$

which gives the same linear current as the semiclassical theory in both AC and DC regimes. Furthermore, by substituting Eq.(C22) into Eq.(C18), where η is also replaced by $\bar{\eta}$, we find

$$\begin{aligned} \rho_{mn}^{(2)} = & \frac{-\delta_{mn} \partial_{\alpha\beta}^2 f_m}{[\omega_1 + i(\eta + \xi)][\omega_\Sigma + i(\eta + 2\xi)]} E_\alpha^{\omega_1} E_\beta^{\omega_2} + \frac{1}{\omega_\Sigma - \epsilon_{mn} + i2\xi} \mathcal{D}_\beta^{mn} \left(\frac{if_{nm} r_{mn}^\alpha}{\omega_1 - \omega_{mn} + i\xi} \right) E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ & + \frac{1}{\omega_\Sigma - \epsilon_{mn} + i2\xi} \frac{ir_{mn}^\beta \partial_\alpha f_{nm}}{\omega_1 + i(\eta + \xi)} E_\alpha^{\omega_1} E_\beta^{\omega_2} + \sum_l \left(\frac{1}{\omega_\Sigma - \epsilon_{mn} + i(\bar{\eta} + 2\xi)} \right) \left(\frac{r_{ml}^\beta r_{ln}^\alpha f_{nl}}{\omega_1 - \epsilon_{ln} + i\xi} - \frac{f_{lm} r_{ml}^\alpha r_{ln}^\beta}{\omega_1 - \epsilon_{ml} + i\xi} \right) \end{aligned} \quad (C23)$$

where $\omega_\Sigma = \omega_1 + \omega_2$. Straightforwardly, substituting Eq.(C23) into the definition of current density $J_\lambda^{(2)} = \sum_{mn} \int_k \rho_{mn}^{(2)} v_{nm}^\lambda \equiv \sigma_{\lambda\alpha\beta}^{(2)}(\omega_1, \omega_2) E_\alpha^{\omega_1} E_\beta^{\omega_2}$, we find

$$\sigma_{\lambda\alpha\beta}^{(2)}(\omega_1, \omega_2) = \sigma_{\lambda\alpha\beta}^{(2,ee)} + \sigma_{\lambda\alpha\beta}^{(2,ei)} + \sigma_{\lambda\alpha\beta}^{(2,ie)} + \sigma_{\lambda\alpha\beta}^{(2,ii)} \quad (\text{C24})$$

with

$$\sigma_{\lambda\alpha\beta}^{(2,ee)} = \sum_{mnl} \int_k \frac{v_{nm}^\lambda}{\omega_\Sigma - \epsilon_{mn} + i(\bar{\eta} + 2\xi)} \left(\frac{r_{ml}^\beta r_{ln}^\alpha f_{nl}}{\omega_1 - \epsilon_{ln} + i\xi} - \frac{f_{lm} r_{ml}^\alpha r_{ln}^\beta}{\omega_1 - \epsilon_{ml} + i\xi} \right), \quad (\text{C25})$$

$$\sigma_{\lambda\alpha\beta}^{(2,ei)} = \sum_{mn} \int_k \left(\frac{v_{nm}^\lambda}{\omega_\Sigma - \epsilon_{mn} + i2\xi} \right) \left(\frac{i r_{mn}^\beta \partial_\alpha f_{nm}}{\omega_1 + i(\eta + \xi)} \right), \quad (\text{C26})$$

$$\sigma_{\lambda\alpha\beta}^{(2,ie)} = \sum_{mn} \int_k \left(\frac{v_{nm}^\lambda}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} \right) \mathcal{D}_\beta^{mn} \left(\frac{i f_{nm} r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} \right), \quad (\text{C27})$$

$$\sigma_{\lambda\alpha\beta}^{(2,ii)} = \sum_n \int_k \frac{-v_n^\lambda \partial_{\alpha\beta}^2 f_n}{[\omega_1 + i(\eta + \xi)] [\omega_\Sigma + i(\eta + 2\xi)]}, \quad (\text{C28})$$

as given by Eqs. (107-110) in the main text.

Appendix D: Calculation details in Section IV

1. Eq.(79)

From Eq.(57), when $m = n$ we find

$$\begin{aligned} \sigma_{\lambda\alpha\beta}^{(2,ee)}(m=n) &\equiv \sum_{nl} \int_k \frac{v_n^\lambda}{(\omega_\Sigma + i2\xi)} \left(\frac{r_{nl}^\beta r_{ln}^\alpha f_{nl}}{\omega_1 - \epsilon_{ln} + i\xi} - \frac{f_{ln} r_{nl}^\alpha r_{ln}^\beta}{\omega_1 - \epsilon_{nl} + i\xi} \right) \\ &= \sum_{nm} \int_k \frac{v_n^\lambda f_{nm}}{(\omega_\Sigma + i2\xi)} \left(\frac{r_{nm}^\beta r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} + \frac{r_{nm}^\alpha r_{mn}^\beta}{\omega_1 - \epsilon_{nm} + i\xi} \right) \\ &= \frac{1}{2} \sum_{nm} \int_k \frac{v_n^\lambda f_{nm}}{(\omega_\Sigma + i2\xi)} \left(\frac{r_{nm}^\beta r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} + \frac{r_{nm}^\alpha r_{mn}^\beta}{\omega_1 - \epsilon_{nm} + i\xi} + \frac{r_{nm}^\alpha r_{mn}^\beta}{\omega_2 - \epsilon_{mn} + i\xi} + \frac{r_{nm}^\beta r_{mn}^\alpha}{\omega_2 - \epsilon_{nm} + i\xi} \right) \\ &= \frac{1}{2} \sum_{nm} \int_k \left[\frac{v_n^\lambda f_{nm} r_{nm}^\beta r_{mn}^\alpha}{(\omega_1 - \epsilon_{mn} + i\xi)(\omega_2 + \epsilon_{mn} + i\xi)} + \frac{v_n^\lambda f_{nm} r_{nm}^\alpha r_{mn}^\beta}{(\omega_1 + \epsilon_{mn} + i\xi)(\omega_2 - \epsilon_{mn} + i\xi)} \right] \\ &= \frac{1}{2} \sum_{nm} \int_k \frac{\Delta_{nm}^\lambda f_{nm} r_{nm}^\beta r_{mn}^\alpha}{(\omega_1 - \epsilon_{mn} + i\xi)(\omega_2 + \epsilon_{mn} + i\xi)} \equiv \sigma_{\lambda\alpha\beta}^{(2,ee1)}, \end{aligned} \quad (\text{D1})$$

where the third line is obtained by symmetrizing (α, ω_1) and (β, ω_2) and $\Delta_{nm}^\lambda = v_n^\lambda - v_m^\lambda$ in the final result. In addition, from the third line of Eq.(D1) to the fourth line, we have used the following relation to remove the divergence,

$$\frac{1}{\omega_1 - \epsilon_{nm} + i\xi} + \frac{1}{\omega_2 - \epsilon_{mn} + i\xi} = \frac{\omega_\Sigma + 2i\xi}{(\omega_1 - \epsilon_{mn} + i\xi)(\omega_2 + \epsilon_{mn} + i\xi)} \quad (\text{D2})$$

So Eq.(D1) is the same as Eq.(79) in the main text.

2. Eq.(94)

To prove Eq.(94), we first define $J_\lambda^{(2;0)} = D + A$, where

$$\begin{aligned} D &\equiv \sum_n \int_k f_n \left(v_n^{(2;\lambda)} + \Omega_n^{(1;\lambda\alpha)} E_\alpha^{\omega_1} \right) = \sum_n \int_k f_n \left(\frac{1}{2} \partial_\lambda a_n^{(1;\alpha)} - \partial_\alpha a_n^{(1;\lambda)} \right) E_\alpha^{\omega_1} \\ &= - \sum_{mn} \int_k \left(\frac{\partial_\lambda f_{nm} r_{nm}^\beta r_{mn}^\alpha}{2(\omega_1 - \epsilon_{mn} + i\xi)} - \frac{\partial_\alpha f_{nm} r_{nm}^\lambda r_{mn}^\beta}{\omega_2 - \epsilon_{mn} + i\xi} \right) E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ &= - \sum_{mn} \int_k \left(\frac{\partial_\lambda f_{nm} r_{nm}^\beta r_{mn}^\alpha}{2(\omega_1 - \epsilon_{mn} + i\xi)} - \frac{\partial_\beta f_{nm} r_{nm}^\lambda r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} \right) E_\alpha^{\omega_1} E_\beta^{\omega_2}, \end{aligned} \quad (D3)$$

which restores Eq.(76) in the DC limit after symmetrizing α and β , as expected. In addition, we have

$$A \equiv \sum_n \int_k f_n \partial_t a_n^{(2;\lambda)} = A_1 + A_2 + A_3 \quad (D4)$$

with

$$A_1 = \text{Re} \sum_{\omega_1 \omega_2} \sum_{nml} \int_k f_n \frac{-2i(\omega_\Sigma + 2i\xi) r_{nm}^\lambda r_{ml}^\beta r_{ln}^\alpha E_\alpha^{\omega_1} E_\beta^{\omega_2}}{(\omega_1 - \epsilon_{ln} + i\xi)(\omega_\Sigma - \epsilon_{mn} + 2i\xi)} + \sum_{\omega_1 \omega_2} \sum_{nml} \int_k f_n \frac{i(\omega_\Sigma + 2i\xi) r_{nl}^\beta r_{lm}^\lambda r_{mn}^\alpha E_\alpha^{\omega_1} E_\beta^{\omega_2}}{(\omega_2 - \epsilon_{nl} + i\xi)(\omega_1 - \epsilon_{mn} + i\xi)} \quad (D5)$$

$$A_2 = \text{Re} \sum_{\omega_1 \omega_2} \sum_{nm} \int_k f_n \frac{2(\omega_\Sigma + 2i\xi) r_{nm}^\lambda \mathcal{D}_{mn}^\beta \left(\frac{r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} \right)}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} E_\alpha^{\omega_1} E_\beta^{\omega_2} \quad (D6)$$

$$A_3 = \text{Re} \sum_{\omega_1 \omega_2} \sum_{nm} \int_k f_n \frac{-(\omega_\Sigma + 2i\xi) r_{nm}^\beta \mathcal{D}_{mn}^\lambda \left(\frac{r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} \right)}{\omega_2 - \epsilon_{nm} + i\xi} E_\alpha^{\omega_1} E_\beta^{\omega_2}, \quad (D7)$$

where we have used Eq.(29) and $\partial_t(E_\alpha^{\omega_1} E_\beta^{\omega_2}) = -i(\omega_\Sigma + i2\xi) E_\alpha^{\omega_1} E_\beta^{\omega_2}$. Note that A_i with $i = 1, 2, 3$ vanishes in the DC limit.

For A_1 , using

$$\frac{\omega_\Sigma + 2i\xi}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} = 1 - \frac{\epsilon_{nm}}{\omega_\Sigma - \epsilon_{mn} + 2i\xi}, \quad (D8)$$

and

$$\frac{\omega_\Sigma + i2\xi}{(\omega_2 - \epsilon_{nl} + i\xi)(\omega_1 - \epsilon_{mn} + i\xi)} = \frac{1}{\omega_2 - \epsilon_{nl} + i\xi} + \frac{1}{\omega_1 - \epsilon_{mn} + i\xi} - \frac{\epsilon_{lm}}{(\omega_2 - \epsilon_{nl} + i\xi)(\omega_1 - \epsilon_{mn} + i\xi)} \quad (D9)$$

we find

$$\begin{aligned} A_1 &= 2\text{Re} \sum_{nml} \int_k f_n \left[\frac{-ir_{nm}^\lambda r_{ml}^\beta r_{ln}^\alpha}{\omega_1 - \epsilon_{ln} + i\xi} + \frac{i\epsilon_{nm} r_{nm}^\lambda r_{ml}^\beta r_{ln}^\alpha}{(\omega_1 - \epsilon_{ln} + i\xi)(\omega_\Sigma - \epsilon_{mn} + 2i\xi)} \right] E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ &\quad + \sum_{nml} \int_k f_n \left[\frac{ir_{nl}^\beta r_{lm}^\lambda r_{mn}^\alpha}{\omega_2 - \epsilon_{nl} + i\xi} + \frac{ir_{nl}^\beta r_{lm}^\lambda r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} - \frac{i\epsilon_{lm} r_{nl}^\beta r_{lm}^\lambda r_{mn}^\alpha}{(\omega_2 - \epsilon_{nl} + i\xi)(\omega_1 - \epsilon_{mn} + i\xi)} \right] E_\alpha^{\omega_1} E_\beta^{\omega_2}. \end{aligned} \quad (D10)$$

Here the first term of the first line and the first two terms of the second line (denoted as A_1^1) can be simplified as

$$\begin{aligned} A_1^1 &= \sum_{\omega_1 \omega_2} \sum_{nml} \int_k f_n \left[\frac{-ir_{nm}^\lambda r_{ml}^\beta r_{ln}^\alpha}{\omega_1 - \epsilon_{ln} + i\xi} + \frac{+ir_{mn}^\lambda r_{lm}^\beta r_{nl}^\alpha}{\omega_1 - \epsilon_{ln} - i\xi} + \frac{ir_{nl}^\beta r_{lm}^\lambda r_{mn}^\alpha}{\omega_2 - \epsilon_{nl} + i\xi} + \frac{ir_{nl}^\beta r_{lm}^\lambda r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} \right] E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ &= \sum_{\omega_1 \omega_2} \sum_{nml} \int_k \frac{ir_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} \left[-f_n r_{nl}^\lambda r_{lm}^\beta - f_m r_{nl}^\beta r_{lm}^\lambda + f_m r_{nl}^\lambda r_{lm}^\beta + f_n r_{nl}^\beta r_{lm}^\lambda \right] E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ &= \sum_{\omega_1 \omega_2} \sum_{nm} \int_k \frac{ir_{mn}^\alpha f_{nm}}{\omega_1 - \epsilon_{mn} + i\xi} \sum_l \left(r_{nl}^\beta r_{lm}^\lambda - r_{nl}^\lambda r_{lm}^\beta \right) E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ &= \sum_{\omega_1 \omega_2} \sum_{nm} \int_k \frac{ir_{mn}^\alpha f_{nm}}{\omega_1 - \epsilon_{mn} + i\xi} (\mathcal{D}_{nm}^\beta r_{nm}^\lambda - \mathcal{D}_{nm}^\lambda r_{nm}^\beta) E_\alpha^{\omega_1} E_\beta^{\omega_2}. \end{aligned} \quad (D11)$$

Note that to obtain the second and third terms before interchanging the dummy indices we have used the $\sum_{\omega_1=\pm\omega} f(\omega_1) = \sum_{\omega_1=\pm\omega} f(-\omega_1)$ and interchanged (α, ω_1) and (β, ω_2) , respectively. And Eq.(83) is used to obtain the final line. For the remaining two terms of in Eq.(D10) (denoted as A_1^2), we find

$$A_1^2 = \sum_{\omega_1\omega_2} \sum_{nml}^{n \neq m} \int_k f_n \left[\frac{v_{nm}^\lambda r_{ml}^\beta r_{ln}^\alpha}{(\omega_1 - \epsilon_{ln} + i\xi)(\omega_\Sigma - \epsilon_{mn} + 2i\xi)} + \frac{v_{mn}^\lambda r_{lm}^\beta r_{nl}^\alpha}{(\omega_1 - \epsilon_{ln} - i\xi)(\omega_\Sigma - \epsilon_{mn} - 2i\xi)} \right] E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ - \sum_{\omega_1\omega_2} \sum_{nml}^{l \neq m} \int_k f_n \frac{r_{nl}^\beta v_{lm}^\lambda r_{mn}^\alpha}{(\omega_2 - \epsilon_{nl} + i\xi)(\omega_1 - \epsilon_{mn} + i\xi)} E_\alpha^{\omega_1} E_\beta^{\omega_2}, \quad (D12)$$

where we have used $v_{nm}^\lambda = i\epsilon_{nm} r_{nm}^\lambda$. By interchanging l and n of the last term and using

$$\frac{1}{(\omega_2 - \epsilon_{ln} + i\xi)(\omega_1 - \epsilon_{ml} + i\xi)} = \frac{1}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} \left(\frac{1}{\omega_1 - \epsilon_{ml} + i\xi} + \frac{1}{\omega_2 - \epsilon_{ln} + i\xi} \right), \quad (D13)$$

we further obtain

$$A_1^2 = \sum_{\omega_1\omega_2} \sum_{nml}^{n \neq m} \int_k \frac{v_{nm}^\lambda}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} \left(\frac{f_n r_{ml}^\beta r_{ln}^\alpha}{\omega_1 - \epsilon_{ln} + i\xi} + \frac{f_m r_{ln}^\beta r_{ml}^\alpha}{\omega_1 + \epsilon_{lm} + i\xi} \right) E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ - \sum_{\omega_1\omega_2} \sum_{nml}^{n \neq m} \int_k \frac{v_{nm}^\lambda}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} \left(\frac{f_l r_{ln}^\beta r_{ml}^\alpha}{\omega_1 - \epsilon_{ml} + i\xi} + \frac{f_l r_{ln}^\beta r_{ml}^\alpha}{\omega_2 - \epsilon_{ln} + i\xi} \right) E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ = \sum_{\omega_1\omega_2} \sum_{nml}^{n \neq m} \int_k \frac{v_{nm}^\lambda}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} \left(\frac{f_{nl} r_{ml}^\beta r_{ln}^\alpha}{\omega_1 - \epsilon_{ln} + i\xi} + \frac{f_{ml} r_{ml}^\alpha r_{ln}^\beta}{\omega_1 - \epsilon_{ml} + i\xi} \right) E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ = \sum_{\omega_1\omega_2} \sigma_{\lambda\alpha\beta}^{(2;ee)} (m \neq n) E_\alpha^{\omega_1} E_\beta^{\omega_2}, \quad (D14)$$

where $\sigma_{\lambda\alpha\beta}^{(2;ee)}$ is defined in Eq.(57). As a result, we find

$$A_1 = A_1^2 + A_1^2 = \sum_{\omega_1\omega_2} \sum_{nm} \int_k \frac{ir_{mn}^\alpha f_{nm}}{\omega_1 - \epsilon_{mn} + i\xi} (\mathcal{D}_{nm}^\beta r_{nm}^\lambda - \mathcal{D}_{nm}^\lambda r_{nm}^\beta) E_\alpha^{\omega_1} E_\beta^{\omega_2} + \sum_{\omega_1\omega_2} \sigma_{\lambda\alpha\beta}^{(2;ee)} (m \neq n) E_\alpha^{\omega_1} E_\beta^{\omega_2}. \quad (D15)$$

For A_2 , we find

$$A_2 = \sum_{\omega_1\omega_2} \sum_{nm} \int_k f_n \left[\frac{(\omega_\Sigma + 2i\xi) r_{nm}^\lambda}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} \mathcal{D}_{mn}^\beta \left(\frac{r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} \right) + \frac{(\omega_\Sigma - 2i\xi) r_{mn}^\lambda}{\omega_\Sigma - \epsilon_{mn} - 2i\xi} \mathcal{D}_{nm}^\beta \left(\frac{r_{nm}^\alpha}{\omega_1 - \epsilon_{mn} - i\xi} \right) \right] E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ = \sum_{\omega_1\omega_2} \sum_{nm} \int_k f_{nm} \frac{(\omega_\Sigma + 2i\xi) r_{nm}^\lambda}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} \mathcal{D}_{mn}^\beta \left(\frac{r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} \right) \quad (D16)$$

where we have used the relation $\sum_{\omega_1=\pm\omega} \sum_{\omega_2=\pm\omega} f(\omega_1, \omega_2) = \sum_{\omega_1=\pm\omega} \sum_{\omega_2=\pm\omega} f(-\omega_1, -\omega_2)$ for the second term. Furthermore, using Eq.(D8), we find

$$A_2 = \sum_{\omega_1\omega_2} \sum_{nm} \int_k \left(-\frac{r_{nm}^\lambda r_{mn}^\alpha \partial_\beta f_{nm}}{\omega_1 - \epsilon_{mn} + i\xi} - \frac{r_{mn}^\alpha f_{nm}}{\omega_1 - \epsilon_{mn} + i\xi} \mathcal{D}_{nm}^\beta r_{nm}^\lambda \right) E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ + \sum_{\omega_1\omega_2} \sum_{nm} \int_k \left[\frac{v_{nm}^\lambda}{\omega_\Sigma - \epsilon_{mn} + 2i\xi} \mathcal{D}_{mn}^\beta \left(\frac{if_{nm} r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} \right) - \frac{iv_{nm}^\lambda r_{mn}^\alpha \partial_\beta f_{nm}}{(\omega_\Sigma - \epsilon_{mn} + 2i\xi)(\omega_1 - \epsilon_{mn} + i\xi)} \right] E_\alpha^{\omega_1} E_\beta^{\omega_2} \\ = \sum_{\omega_1\omega_2} \sum_{nm} \int_k \left(-\frac{r_{nm}^\lambda r_{mn}^\alpha \partial_\beta f_{nm}}{\omega_1 - \epsilon_{mn} + i\xi} - \frac{r_{mn}^\alpha f_{nm}}{\omega_1 - \epsilon_{mn} + i\xi} \mathcal{D}_{nm}^\beta r_{nm}^\lambda \right) E_\alpha^{\omega_1} E_\beta^{\omega_2} + \left(\sigma_{\lambda\alpha\beta}^{(2;ie)} + \sigma_{\lambda\alpha\beta}^{(ei;0)} \right) E_\alpha^{\omega_1} E_\beta^{\omega_2}, \quad (D17)$$

where we have used Eq.(59) and Eq.(91).

For A_3 , using integration by parts we find:

$$\begin{aligned}
A_3 &= \text{Re} \sum_{\omega_1 \omega_2} \sum_{nm} \int_k \left[\frac{(\omega_\Sigma + 2i\xi) f_n r_{mn}^\alpha \mathcal{D}_{nm}^\lambda r_{nm}^\beta}{(\omega_2 - \epsilon_{nm} + i\xi)(\omega_1 - \epsilon_{mn} + i\xi)} + \frac{r_{nm}^\beta r_{mn}^\alpha}{\omega_1 - \epsilon_{mn} + i\xi} \partial_\lambda \left(\frac{(\omega_\Sigma + 2i\xi) f_n}{\omega_2 - \epsilon_{nm} + i\xi} \right) \right] E_\alpha^{\omega_1} E_\beta^{\omega_2} \\
&= \text{Re} \sum_{\omega_1 \omega_2} \sum_{nm} \int_k \left[\frac{r_{mn}^\alpha \mathcal{D}_{nm}^\lambda r_{nm}^\beta}{\omega_1 - \epsilon_{mn} + i\xi} + \frac{r_{mn}^\alpha \mathcal{D}_{nm}^\lambda r_{nm}^\beta}{\omega_2 - \epsilon_{nm} + i\xi} \right] f_n E_\alpha^{\omega_1} E_\beta^{\omega_2} \\
&\quad + \text{Re} \sum_{\omega_1 \omega_2} \sum_{nm} \int_k \left[\frac{r_{mn}^\alpha r_{nm}^\beta}{\omega_1 - \epsilon_{mn} + i\xi} + \frac{r_{mn}^\alpha r_{nm}^\beta}{\omega_2 - \epsilon_{nm} + i\xi} \right] \partial_\lambda f_n E_\alpha^{\omega_1} E_\beta^{\omega_2} \\
&\quad + \text{Re} \sum_{\omega_1 \omega_2} \sum_{nm} \int_k \left[\frac{r_{nm}^\beta r_{mn}^\alpha \Delta_{nm}^\lambda}{(\omega_1 - \epsilon_{mn} + i\xi)(\omega_2 - \epsilon_{nm} + i\xi)} + \frac{r_{nm}^\beta r_{mn}^\alpha \Delta_{nm}^\lambda}{(\omega_2 - \epsilon_{nm} + i\xi)^2} \right] f_n E_\alpha^{\omega_1} E_\beta^{\omega_2} \\
&= \sum_{\omega_1 \omega_2} \sum_{mn} \int_k \frac{f_{nm} r_{mn}^\alpha \mathcal{D}_{nm}^\lambda r_{nm}^\beta}{\omega_1 - \epsilon_{mn} + i\xi} E_\alpha^{\omega_1} E_\beta^{\omega_2} + \frac{1}{2} \sum_{\omega_1 \omega_2} \sum_{mn} \int_k \frac{\partial_\lambda f_{nm} r_{mn}^\alpha r_{nm}^\beta}{\omega_1 - \epsilon_{mn} + i\xi} E_\alpha^{\omega_1} E_\beta^{\omega_2} + \sum_{\omega_1 \omega_2} \sigma_{\lambda\alpha\beta}^{(2;ee)}(m=n) E_\alpha^{\omega_1} E_\beta^{\omega_2},
\end{aligned} \tag{D18}$$

$$\tag{D19}$$

where we have used Eq.(79), which is derived in Appendix D 1. In addition, we have used the following relations

$$\frac{\omega_\Sigma + 2i\xi}{(\omega_1 - \epsilon_{nm} + i\xi)(\omega_2 - \epsilon_{mn} + i\xi)} = \frac{1}{\omega_2 - \epsilon_{mn} + i\xi} + \frac{1}{\omega_1 - \epsilon_{nm} + i\xi} \tag{D20}$$

$$\frac{\Delta_{nm}^\lambda}{(\omega_2 - \epsilon_{nm} + i\xi)^2} = \partial_\lambda \left(\frac{1}{\omega_2 - \epsilon_{nm} + i\xi} \right), \quad \partial_\lambda (r_{nm}^\beta r_{mn}^\alpha) = \mathcal{D}_{nm}^\lambda (r_{nm}^\beta) r_{mn}^\alpha + r_{nm}^\beta (\mathcal{D}_{mn}^\lambda r_{mn}^\alpha) \tag{D21}$$

to obtain the last line.

Finally, combining Eq.(D3), Eq.(D15), Eq.(D17) with Eq.(D21), we find

$$\begin{aligned}
J_\lambda^{(2;0)} &= D + A_1 + A_2 + A_3 = \sum_{\omega_1 \omega_2} \left[\sigma_{\lambda\alpha\beta}^{(2;ee)}(m=n) + \sigma_{\lambda\alpha\beta}^{(2;ee)}(m \neq n) + \sigma_{\lambda\alpha\beta}^{(2;ie)} + \sigma_{\lambda\alpha\beta}^{(ie;0)} \right] E_\alpha^{\omega_1} E_\beta^{\omega_2} \\
&= \sum_{\omega_1 \omega_2} \left[\sigma_{\lambda\alpha\beta}^{(2;ee)} + \sigma_{\lambda\alpha\beta}^{(2;ie)} + \sigma_{\lambda\alpha\beta}^{(ie;0)} \right] E_\alpha^{\omega_1} E_\beta^{\omega_2},
\end{aligned} \tag{D22}$$

which gives the Eq.(D 2) in the main text.

Appendix E: The energy with the Kohn-Luttinger method

The Hamiltonian $H = H_0 + H_1$ satisfies the time-dependent Schrödinger equation

$$i\partial_t |\psi\rangle = H |\psi\rangle. \tag{E1}$$

Performing a unitary transformation for the quantum state: $|\psi\rangle \rightarrow e^S |\psi\rangle \equiv |\psi'\rangle$, we find:

$$i\partial_t |\psi'\rangle = (i\partial_t e^S) |\psi\rangle + e^S i\partial_t |\psi\rangle = (i\partial_t e^S) e^{-S} |\psi'\rangle + e^S H e^{-S} |\psi'\rangle \equiv H' |\psi'\rangle, \tag{E2}$$

where we have used Eq. (E1) and the Hamiltonian after this unitary transformation (denoted as H') is explicitly given by:

$$H \rightarrow H' \equiv (i\partial_t e^S) e^{-S} + e^S H e^{-S} = e^S (-i\partial_t e^{-S}) + e^S H e^{-S} \tag{E3}$$

where we have used that $0 = \partial_t (e^S e^{-S}) = (\partial_t e^S) e^{-S} + e^S (\partial_t e^{-S})$. Furthermore, using the operator identity:

$$e^A B e^{-A} = B + \frac{1}{1!} [A, B] + \frac{1}{2!} [A, [A, B]] + \frac{1}{3!} [A, [A, [A, B]]] + \dots, \tag{E4}$$

we find

$$\begin{aligned}
H' &= \left(1 + S + \frac{1}{2}S^2 + \dots\right) (-i\partial_t) \left(1 - S + \frac{1}{2}S^2 + \dots\right) + \left(H + [S, H] + \frac{1}{2}[S, [S, H]] + \dots\right) \\
&= \left(i\partial_t S + Si\partial_t S - \frac{1}{2}i\partial_t S^2\right) + \left(H + [S, H] + \frac{1}{2}[S, [S, H]]\right) \\
&= i\partial_t S + \frac{1}{2}[S, i\partial_t S] + H_0 + H_1 + [S, H_0] + [S, H_1] + \frac{1}{2}[S, [S, H]] \\
&= H_0 + [S, H_1] + \frac{1}{2}[S, [S, H] + i\partial_t S]
\end{aligned} \tag{E5}$$

where we only keep terms to the order of S^2 and choose

$$i\partial_t S + H_1 + [S, H_0] = 0 \tag{E6}$$

to fix the unitary transformation S . We remark that this gauge choice bypasses the first-order energy correction in H' . At this stage, by using the basis of $H_0|n\rangle = \epsilon_n|n\rangle$, we further find that

$$\langle n | (i\partial_t S + H_1 + [S, H_0]) | m \rangle = 0 \implies \partial_t S_{nm} + i\epsilon_{nm} S_{nm} = iE_\alpha(t) r_{nm}^\alpha, \quad (m \neq n) \tag{E7}$$

where $E_\alpha(t) = \sum_{\omega_\alpha} E_\alpha e^{-i\omega_\alpha t}$ with $\omega_\alpha = \pm\omega$ and r_{nm}^α is the interband Berry connection. By solving Eq.(E7), we obtain:

$$S_{nm} = e^{-i\epsilon_{nm}t} \left(\sum_{\omega_\alpha} iE_\alpha r_{nm}^\alpha \int e^{-i\omega_\alpha t} e^{\int i\epsilon_{nm} dt} dt \right) = - \sum_{\omega_\alpha} \frac{r_{nm}^\alpha E_\alpha e^{-i\omega_\alpha t}}{\omega_\alpha - \epsilon_{nm}}. \tag{E8}$$

As a result, the energy for H' can be calculated as:

$$\begin{aligned}
E_n &= \langle n | H' | n \rangle = \epsilon_n + \langle n | [S, H_1] | n \rangle + \frac{1}{2} \langle n | [S, [S, H_0] + i\partial_t S] | n \rangle \\
&= \epsilon_n + \langle n | [S, H_1] | n \rangle - \frac{1}{2} \langle n | [S, H_1] | n \rangle \\
&= \epsilon_n + \frac{1}{2} \langle n | [S, H_1] | n \rangle \\
&= \epsilon_n + \frac{1}{2} \sum_{\omega_\alpha} \sum_{\omega_\beta} \sum_m \left(-\frac{E_\alpha E_\beta r_{nm}^\alpha r_{mn}^\beta e^{-i(\omega_\alpha + \omega_\beta)t}}{\omega_\alpha - \epsilon_{nm}} + \frac{E_\alpha E_\beta r_{nm}^\beta r_{mn}^\alpha e^{-i(\omega_\alpha + \omega_\beta)t}}{\omega_\alpha - \epsilon_{mn}} \right).
\end{aligned} \tag{E9}$$

Note that we have used Eq.(E6) and dropped $\frac{1}{2} \langle n | [S, [S, H_1]] | n \rangle \propto E_\alpha^3$. Under the DC limit ($\omega_\alpha \rightarrow 0$), we obtain:

$$E_n = \epsilon_n + \frac{1}{2} \sum_m \frac{g_{nm}^{\alpha\beta} E_\alpha E_\beta}{\epsilon_{nm}}, \tag{E10}$$

as given by Eq. (98) in the main text. Instead of directly taking the DC limit, if we first perform the frequency summation:

$$\begin{aligned}
E_n &= \epsilon_n + \frac{1}{2} \sum_m \left(-\frac{E_\alpha E_\beta r_{nm}^\alpha r_{mn}^\beta e^{-i2\omega t}}{\omega - \epsilon_{nm}} + \frac{E_\alpha E_\beta r_{nm}^\beta r_{mn}^\alpha e^{-i2\omega t}}{\omega - \epsilon_{mn}} \right) \\
&\quad + \frac{1}{2} \sum_m \left(-\frac{E_\alpha E_\beta r_{nm}^\alpha r_{mn}^\beta}{\omega - \epsilon_{nm}} + \frac{E_\alpha E_\beta r_{nm}^\beta r_{mn}^\alpha}{\omega - \epsilon_{mn}} \right) \\
&\quad + \frac{1}{2} \sum_m \left(-\frac{E_\alpha E_\beta r_{nm}^\alpha r_{mn}^\beta}{-\omega - \epsilon_{nm}} + \frac{E_\alpha E_\beta r_{nm}^\beta r_{mn}^\alpha}{-\omega - \epsilon_{mn}} \right) \\
&\quad + \frac{1}{2} \sum_m \left(-\frac{E_\alpha E_\beta r_{nm}^\alpha r_{mn}^\beta e^{+i2\omega t}}{-\omega - \epsilon_{nm}} + \frac{E_\alpha E_\beta r_{nm}^\beta r_{mn}^\alpha e^{+i2\omega t}}{-\omega - \epsilon_{mn}} \right).
\end{aligned} \tag{E11}$$

Under the DC limit, we obtain

$$E_n = \epsilon_n + 2 \sum_m \frac{g_{nm}^{\alpha\beta} E_\alpha E_\beta}{\epsilon_{nm}}, \quad (\text{E12})$$

as given in Ref. [69], where the additional factor 2 arises from the summation over two frequencies (Note that the electric field appears twice) but Ref. [69] only considers one. In the main text, the DC limit is directly taken without considering the frequency summation.

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