Zero frequency divergence and gauge phase factor in the optical response theory

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The static current-current correlation leads to the definitonal zero frequency divergence (ZFD) in the optical susceptibilities. Previous computations have shown nonequivalent results between two gauges (p \cdot A and E \cdot r) under the exact same unperturbed wave functions. We reveal that those problems are caused by the improper treatment of the time-dependent gauge phase factor in the optical response theory. The gauge phase factor, which is conventionally ignored by the theory, is important in solving ZFD and obtaining the equivalent results between these two gauges. The Hamiltonians with these two gauges are not necessarily equivalent unless the gauge phase factor is properly considered in the wavefunctions. Both Su-Shrier-Heeger (SSH) and Takayama-Lin-Liu-Maki (TLM) models of trans-polyacetylene serve as our illustrative examples to study the linear susceptibility \( \chi^{(1)} \) through both current-current and dipole-dipole correlations. Previous improper results of the \( \chi^{(1)} \) calculations and distribution functions with both gauges are discussed. The importance of gauge phase factor to solve the ZFD problem is emphasized based on SSH and TLM models. As a conclusion, the reason why dipole-dipole correlation favors over current-current correlation in the practical computations is explained.

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I. INTRODUCTION

The static current-current correlation (\( J_0 J_0 \)) is widely applied in the optical response theory for many decades, both in the definition of linear susceptibility \( \chi^{(1)} \) or conductivity \( \sigma^{(1)} \), and in the definition of the nonlinear optical susceptibilities such as \( \chi^{(n)} \), where \( n \geq 2 \).

Within the semiclassical theory of radiation, which is also literaturely emphasized by Mahan, Bucher and Cotter, Bloembergen, Shen, and Mukamel, the electric field are treated classically and propagation of electromagnetic waves in a medium is governed by Maxwell’s equations, that is, the electric field \( \mathbf{E}(\mathbf{r}, t) \) at some specific position \( \mathbf{r} \) and time \( t \) can be described as:

\[
\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_0 e^{i\mathbf{k} \cdot \mathbf{r} - i\omega t},
\]

where \( \mathbf{E}_0 \) is the amplitude, \( \mathbf{k} \) and \( \omega \) are the wave vector and frequency. Then, the \( \chi^{(n)} \) in a unit volume \( v \) under the static current-current (\( J_0 J_0 \)) correlation is conventionally defined as follows:

\[
\chi^{(n)}(\mathbf{r}_1, \ldots, \mathbf{r}_n; \omega_1, \ldots, \omega_n) = -\delta_{n,1} n^{(e)} e^2 \hat{I} + \frac{\chi^{(1)}(\mathbf{r}_1, \ldots, \mathbf{r}_n; \omega_1, \ldots, \omega_n)}{\epsilon_0 i \Omega \omega_1 \cdots \omega_n},
\]

where \( \Omega \equiv -\sum_{i=1}^{n} \omega_i \), \( n^{(e)} \) and \( m \) are the electron density and electron mass, \( \epsilon_0 \) the dielectric constant, \( \hat{I} \) a unit dyadic, \( \delta_{n,1} \) Kronecker symbol, and

\[
\chi^{(n)}_{j_0 j_0}(\mathbf{r}_1, \ldots, \mathbf{r}_n; \omega_1, \ldots, \omega_n) = \frac{1}{n!} \left[ \frac{1}{\hbar} \right]^n \int d\mathbf{r}_1 \cdots d\mathbf{r}_n \int dt_1 \cdots dt_n \int d\mathbf{r} dt e^{-i\mathbf{k} \cdot \mathbf{r} + i\Omega t} \langle \hat{T} \hat{J}_0(\mathbf{r}, t) \hat{J}_0(\mathbf{r}_1, t_1) \cdots \hat{J}_0(\mathbf{r}_n, t_n) \rangle,
\]

with \( \hat{T} \) is the time-ordering operator and \( \hat{J}_0 \) is the static current operator.
If we choose the static dipole-dipole ($DD$) correlation, the $n$th-order susceptibilities will be obtained as follows:

\[
\chi^{(n)}(\Omega; \omega_1, \ldots, \omega_n) = \frac{1}{n!} \left[ \frac{i}{\hbar} \right]^n \int dr_1 \cdots dr_n \int dt_1 \cdots dt_n \int d\mathbf{r} e^{-i\mathbf{k} \cdot \mathbf{r} + i\mathbf{A} \cdot \mathbf{r}} (\hat{T} \mathbf{D}(\mathbf{r}, t) \mathbf{D}(\mathbf{r}_1, t_1) \cdots \mathbf{D}(\mathbf{r}_n, t_n)),
\]

(1.4)

where $\mathbf{D}$ is the static dipole operator.

It is commonly held that the apparent zero frequency divergence (ZFD) through the static current expression Eq.(1.2) is only a virtual problem, and the gauge $\mathbf{E} \cdot \mathbf{r}$ and $\mathbf{p} \cdot \mathbf{A}$ will give the exactly same results with the same unperturbed wave functions. In other words, Eq.(1.2) and Eq.(1.4) should reach the same result if one proceeds properly. In linear response theory, for the homogeneous and isotropic medium, Martin and Schwinger have shown that the ZFD in conductivity $\sigma^{(1)}$ through current expressions could be cancelled by introducing diamagnetic term ($A^2$ term). The cancellation of ZFD in the linear conductivity by diamagnetic term is also discussed by Mahan, Haug and Jauho in their famous books, with a careful consideration on the limitation sequence between $k$ and $\omega$. In solid state, for either full-filled or empty bands, Aspnes had shown the equivalence of two gauges (from Eq.(1.2) and Eq.(1.4)) based on the assumption of the cancellation of a ZFD term Eq.(2.6) in his seminal work of $\chi^{(2)}$ computations. These works strengthen the common feelings of the equivalence between two gauges under the static dipole or the static current expressions. And the issue of ZFD to be a virtual problem was usually borrowed with limited justification.

Although the ‘virtual’ property of this ZFD are literally emphasized by a lot of people, strictly speaking, as to our knowledge, the solution to this ZFD problem is seldom directly obtained from $J_0 J_0$ correlation [like Eq.(1.2)] except Martin and Schwinger’s original proof. As we can see in the calculation of $\chi^{(2)}$ in solid state, the ZFD term is conventionally isolated from the convergent term and then is discarded without careful direct check. Historically, this ZFD problem is of no interest due to the following facts: (i). The gauge transformations seem to guarantee the equivalence of two gauges under the same set of wave functions; (ii). In transport theory, the correct imaginary part of $J_0 J_0$ correlation ($\text{Re} \sigma^{(1)}(\omega)$) still can be obtained and (iii) the ZFD problem (related to $\text{Im} \sigma^{(1)}(\omega)$) usually can be avoided by applying the Kramers-Kronig (KK) relations on the imaginary part of $J_0 J_0$ correlation. Thus the equivalence of two gauges becomes a well-accepted opinion in the community and the ZFD is widely considered to be at most a complex technical problem.

Besides those assumptions made in the previous ZFD proof, however, in the practical applications, there always exists some puzzles from the above common feelings. In a careful study on the gauge choice of two-photon transitions in 1s-2s transition of hydrogen atom, Bassani, Forney and Quattropani have found that by directly applying the exact same unperturbed wave functions, $\mathbf{E} \cdot \mathbf{r}$ converges much faster than $\mathbf{p} \cdot \mathbf{A}$ by using a limited number of discrete intermediate states; Numerical calculations also show a 50% difference of the transition rate between those gauges if we only included all discrete intermediate states. Thus, Bassani et al. draw the conclusion that $\mathbf{E} \cdot \mathbf{r}$ is a better gauge. While the study of oscillator strength in a superlattice, by Peeters et al., turns out that $\mathbf{E} \cdot \mathbf{r}$ and $\mathbf{p} \cdot \mathbf{A}$ is nonequivalent in the barrier region based on numerical computations. Thus, on the contrary, Peeters et al. concluded that position operator $\mathbf{r}$ in the solid state should be redefined and $\mathbf{p} \cdot \mathbf{A}$ is much better. A recent study on zinc-blende semiconductor again brings the questions on the equivalence role of $\mathbf{E} \cdot \mathbf{r}$ and $\mathbf{p} \cdot \mathbf{A}$ in representing the transition matrix of $\chi^{(2)}$ formula.

Extensive studies on the optical properties of polymers are based on the tight-binding approximate (TBA) models, such as Su-Shrieffer-Heeger (SSH) model and Takayama-Lin-Liu-Maki (TLM) model in weakly correlated systems, Hubbard and Pariser-Parr-Pople (PPP) models in strongly correlated systems. Those models drastically reduce the complicity of the systems and provide a reasonable way to reveal the actual physical insights of many-body systems. In providing the gauge invariance, a U(1) phase transformation has been suggested in those models. Based on the static current formula without diamagnetic term, there will be ZFD problem in the linear conductivity $\sigma^{(1)}(\omega)$. However, this ZFD problem has not been pointed out clearly and obviously neglected by the previous works. To avoid ZFD in $\sigma^{(1)}$ and to obtain a convergent result, Batistic and Bishop suggest to subtract the term $\langle j_0 j_0 \rangle(\omega = 0)$, which is supposed to be the diamagnetic term directly derived from Hamiltonian. Unfortunately, as we will show in this paper that the diamagnetic term derived from U(1) transformation can not directly return the expected $\langle j_0 j_0 \rangle(\omega = 0)$ term. Moreover, experimentally observed two-photon absorption peak in the $\chi^{(3)}$ spectrum of trans-polyacetylene has raised wide interest for the theoretical explanation. From Eq.(1.2) based on TLM models, two-photon cusp was obtained analytically, but it has been criticized by the others from the dipole formula approach and other physical concern. Recently, a quite different analytical form of the
$\chi^{(3)}$ spectrum [34] is obtained under $DD$ correlation from that under $J_0J_0$ correlation. Those above discrepancies indeed already cast some doubts on the rooted belief of equivalence between $J_0J_0$ and $DD$ correlations.

In this paper, we will reexamine the concepts of the gauge transformations and directly show that the gauge phase factor, which is not sufficiently emphasized previously and is conventionally ignored under the current-current correlation scheme, is actually very important in the optical response theory to solve this ZFD difficulty, and to recover the equivalent results between the two gauges ($p \cdot A$ and $r \cdot E$). Therefore, the static current operator $J_0$ is no longer suitable for considering the equivalence between two gauges, instead, we should include the induced field currents (IFC) which are introduced by the gauge phase factor. To illustrate the effect of gauge phase factor rather than the abstract concepts, we only choose the linear response under the one-dimensional ($1d$) periodic TBA models like SSH and TLM models as our examples. Certainly the concepts of the gauge phase factor under the specific linear examples also can be expanded into the nonlinear optical response theory and two-dimensional ($2d$) or three-dimensional ($3d$) cases.

The paper is organized as follows: In Sec.II, we will reexamine the concept of the gauge transformation and discuss the importance of the gauge phase factor in the optical response theory. The problems caused by the ignorance of the gauge phase factor are discussed under a general scheme independent of the models. To give an intuitive picture, linear optical response under periodic tight-binding-models (TBA) such as both SSH and TLM models is investigated in this paper, the Hamiltonian under $DD$ correlation ($E \cdot r$) is discussed (Sec.III.A), and we will study the linear susceptibility $\chi^{(1)}$ through $DD$ correlation in Sec.III.B. The qualitative different results by the conventional polarization operator $\hat{P}$ in optical response are discussed based on periodic models in Sec.III.C. $\chi^{(1)}$ under current-current correlation is discussed in Sec.IV. The SSH Hamiltonian of $p \cdot A$ will be obtained in Sec.IV.A. Before applying the gauge phase factor, the ZFD problem of $\chi^{(1)}$ through $J_0J_0$ correlation is illustrated in Sec.IV.B, previous qualitatively different solutions of $\chi^{(1)}$ and the practical difficulties in this ZFD problem based on the models are also analyzed (Sec.IV.B). After applying the gauge phase factor to the wave functions, a convergent result can be obtained and the ZFD problem will be solved (Sec.IV.C).

The conditions of equivalence between two gauges are discussed in Sec.V.A and the influence of gauge phase factor on the initial distribution function $f_{\alpha}(k)$ under two gauges are investigated (Sec.V.B). The reasons of some previous puzzles on the choice of the gauges are discussed in Sec.V.C. The conclusion emphasizing the implications of our work will be described in Sec.VI.

II. GAUGE PHASE FACTOR IN GAUGE TRANSFORMATION

Gauge transformation has already been well-understood in the optical response theory. [37,38] The equivalence between two gauges is built up on the concept of gauge transformation.

If the electromagnetic field applied, the Schrödinger equation is given by:

$$i\hbar \frac{\partial}{\partial t} \psi(r,t) = \left[ \frac{1}{2m}(\vec{p} - qA)^2 + V(r) + q\phi \right] \psi(r,t), \tag{2.1}$$

where $\psi(r,t)$ is the exact wave function at space position $r$ and specific time $t$, $m$ is the particle mass, $q$ is the electrical charge, $V(r)$ is the potential, $A$ and $\phi$ as vector and scalar potential correspondingly, under the following transformation:

$$\begin{cases} A \rightarrow A' = A + \nabla f(r,t) \\ \phi \rightarrow \phi' = \phi - \frac{\partial}{\partial t} f(r,t), \end{cases} \tag{2.2}$$

where $f(r,t)$ is arbitrary, and $A'$ and $\phi'$ are new vector and new scalar potentials after the transformation Eq.(2.2). It could be shown [38] that the form of the Schrödinger equation will be exactly the same if the old wave function $\psi$ makes the following change into the new exact wave function $\psi'$:

$$\psi \rightarrow \psi' = e^{iF_g(r,t)} \psi = \hat{T}_G(r,t) \psi, \tag{2.3}$$

where gauge phase factor $F_g(r,t)$ is defined as:

$$F_g(r,t) = \frac{q}{\hbar} f(r,t). \tag{2.4}$$

The above Eq.(2.3) and Eq.(2.3) are called gauge transformation (or $U(1)$ transformation [24]).
Long-wavelength approximation \[2,3\] is used in this paper, that is, the \( k = 0 \) in Eq.(1.1), and the electric field \( E \) is described as \( E = E_0 e^{-i\omega t} \).

If we consider the following initial scalar and vector potentials under \( \mathbf{E} \cdot \mathbf{r} \) gauge:

\[
A = 0, \phi = -\mathbf{E} \cdot \mathbf{r}.
\]  

(2.5)

After choosing the gauge phase factor \( F_g \) as

\[
F_g = \frac{q \mathbf{E} \cdot \mathbf{r}}{i\hbar \omega},
\]  

(2.6)

by Eq.(2.2), we obtain the new vector and new scalar potential under \( \mathbf{p} \cdot \mathbf{A} \) gauge as:

\[
A' = \frac{\mathbf{E}}{i\omega}, \phi' = 0.
\]  

(2.7)

The connection between the old and new wave function is determined by Eq.(2.3).

Under perturbative schemes to study the optical response, conventionally people use the exact same set of unperturbed wave functions \( \psi^0_n(\mathbf{r}, t) \) of Hamiltonian \( \hat{H}_0 \) (when \( \mathbf{A} = 0 \) and \( \psi = 0 \) in Eq.(2.1)) to serve as our expansion basis for both \( \mathbf{E} \cdot \mathbf{r} \) and \( \mathbf{p} \cdot \mathbf{A} \) gauges. However, we should point out that the wave functions for both \( \mathbf{E} \cdot \mathbf{r} \) and \( \mathbf{p} \cdot \mathbf{A} \) gauges (before and after gauge transformation) should also be restricted by the gauge phase factor \( F_g \) from Eq.(2.3), therefore two basis sets for both gauges are not the exact same unperturbed wave functions \( \psi^0_n(\mathbf{r}, t) \), but are different by the gauge phase factor \( F_g \). And the Hamiltonian under two gauges(\( \mathbf{E} \cdot \mathbf{r} \) and \( \mathbf{p} \cdot \mathbf{A} \)) are not necessary equivalent if they are treated independently and are isolated from the connection between the wave functions under the two gauges. Unfortunately, this crucial point has not been clearly illustrated and obviously missed by the perturbated schemes works. Especially under current-current correlation scheme, the gauge phase factor’s contribution is obviously ignored and \( A^2(t) \) term is considered as no any physical meanings. Thus the current-current correlation is conventionally reduced into the \( J_0 J_0 \) formula such as Eq.(1.2), and the equivalence between current-current and dipole-dipole correlations is usually considered as \( J_0 J_0 \) and \( DD \) correlations under the exact same basis of unperturbed wave functions.

An elegant review by Langhoff, Epstein and Karplus covered the topics of time-dependent perturbative theory, they have sharply pointed out that the time-dependent phase in wave functions is very essential and the improper treatment of time-dependent phase will cause secular divergence in time-dependent perturbations. In field theory, it is also well-understood that the improper treatment of the phase factor will cause divergence. Since the gauge phase factor Eq.(2.6) is obviously time-dependent, neglecting this phase factor will cause the ZFD in the susceptibility computations, as the examples we will show in the following sections.

### III. LINEAR RESPONSE BY DIPOLE-DIPOLE CORRELATION

Having appreciated the importance of the gauge phase factor, we choose the following single electron periodic models – SSH (or Hückel) model and TLM model as our examples provided the following reasons: (i). Those periodic models are widely applied in the polymer theory in the 80s and early 90s; Remarkable results have been obtained; (ii). The optical susceptibilities obtained from those models can be analytically solvable and be compared with the previous results; (iii). In both the SSH and the TLM models, Peierls instability leads to the semiconductor property of two band structure – with valence band full-filled and conduction band empty. It is very obvious from the physical point of view, as the frequency of the electrical field goes 0 (reaches the static electric field), the linear conductivity \( \sigma^{(1)} \) and the linear susceptibility \( \chi^{(1)} \) will not be reduced to Drude formula as in the metals, and will not cause ZFD problem. In this section, we will first discuss \( \chi^{(1)} \) and \( \sigma^{(1)} \) of infinite chains (with number of \( (CH) \) unit \( N \) goes to infinity) under the \( DD \) correlation.

#### A. SSH Hamiltonian

Based on periodic TBA, The SSH Hamiltonian is given by:

\[
H_{SSH} = -\sum_{l,s} \left[ t_0 + (-1)^l \frac{\Delta}{2} \right] (\hat{C}^\dagger_{l+1,s} \hat{C}_{l,s} + \hat{C}^\dagger_{l,s} \hat{C}_{l+1,s}),
\]  

(3.1)
where \( t_0 \) is the transfer integral between the nearest-neighbor sites, \( \Delta \) is the gap parameter and \( \hat{C}^\dagger_{l,s}(\hat{C}_{l,s}) \) creates (annihilates) an \( \pi \) electron at site \( l \) with spin \( s \). In continuum limitation, above SSH model will give the TLM model. \(^{27}\) For the SSH model, each site is occupied by one electron.

If we want to include the electron-photon interaction \( \mathbf{E} \cdot \mathbf{r} \) directly from the polarization operator \( \hat{P} \), where

\[
\hat{P} = \sum_l R_l \hat{C}^\dagger_l \hat{C}_l, \tag{3.2}
\]

and

\[
R_l = l a + (-1)^l u \tag{3.3}
\]

is the site \( l \) position with the lattice constant \( a \) and dimerized constant \( u \), \(^{24}\) we will face the problem of ill-definition of \( \hat{P} \) in periodic systems. \(^{13,17,23}\) To solve this problem, we should consider the imposed periodic condition of the position operator \( \mathbf{r} \). \(^{10,14}\) Expressing position operator \( \mathbf{r} \) under the Bloch states \( |n, \mathbf{k}, \mathbf{r} \rangle = u_{n,k}(\mathbf{r}) e^{i\mathbf{k} \cdot \mathbf{r}}, \) where \( u_{n,k}(\mathbf{r}) \) is the periodic function under the translation of lattice vector, \(^{10}\) we will be able to satisfy the periodic condition of \( \mathbf{r} \) as follows:

\[
r_{n,k,n',k'} = i \delta_{n,n'} \nabla_k \delta(\mathbf{k} - \mathbf{k}') + \Omega_{n,n'}(k) \delta(k - k'), \tag{3.4}
\]

and

\[
\Omega_{n,n'}(k) = \frac{i}{v} \int_v u_{n,k}(\mathbf{r}) \nabla_k u_{n',k}(\mathbf{r}) d\mathbf{r}, \tag{3.5}
\]

where \( v \) is unit cell volume.

We change Hamiltonian Eq.\((3.1)\) into the momentum space by applying the following consecutive transformations:

\[
\left\{ \begin{aligned}
\hat{C}_{l,s} &= \frac{1}{\sqrt{N}} \sum_{-\frac{\pi}{a} \leq k < \frac{\pi}{a}} (\hat{C}^w_{k,s} + \hat{C}^c_{k,s}) e^{ik R_l}, \\
\hat{C}_{k,s} &= \frac{1}{\sqrt{N}} \sum_{-\frac{\pi}{a} \leq k < \frac{\pi}{a}} (\hat{C}^w_{k,s} - \hat{C}^c_{k,s}) e^{ik R_l},
\end{aligned} \right. \tag{3.6}
\]

and

\[
\left\{ \begin{aligned}
\hat{a}^v_{k,s} &= -i\gamma_k \hat{C}^v_{k,s} + \xi_k \hat{C}^c_{k,s}, \\
\hat{a}^c_{k,s} &= i\xi_k \hat{C}^v_{k,s} + \gamma_k \hat{C}^c_{k,s},
\end{aligned} \right. \tag{3.7}
\]

with

\[
\gamma_k = \frac{1}{\sqrt{2}} \sqrt{1 + \frac{2t_0 \cos(ka)}{\varepsilon(k)}},
\]

\[
\xi_k = \frac{sgn(k)}{\sqrt{2}} \sqrt{1 - \frac{2t_0 \cos(ka)}{\varepsilon(k)}}, \tag{3.8}
\]

where

\[
\varepsilon(k) = \sqrt{[2t_0 \cos(ka)]^2 + [\Delta \sin(ka)]^2} \tag{3.9}
\]

and \( R_{l_0} \) and \( R_{l_0} \) are odd and even position defined by Eq.\((3.3)\). \( \hat{a}^v_{k,s}(t) \) and \( \hat{a}^c_{k,s}(t) \) are the excitations of electrons in the conduction band and valence band with momentum \( k \) and spin \( s \).

If we choose the spinor description \( \hat{\psi}^\dagger_{k,s}(t) = (\hat{a}^v_{k,s}(t), \hat{a}^c_{k,s}(t)) \), SSH Hamiltonian including \( \mathbf{E} \cdot \mathbf{r} \) in momentum space is described by:

\[
\hat{H}_{SSH}(k,t) = \hat{H}_0 + \hat{H}_{E \cdot r}, \tag{3.10}
\]
where
\[ \hat{H}_0 = \sum_{-\frac{\pi}{2a} \leq \vec{k} \leq \frac{\pi}{2a}} \epsilon(\vec{k}) \hat{\psi}_{k,s}^\dagger(t) \sigma_3 \hat{\psi}_{k,s}(t) \]  
(3.11)

and
\[ \hat{H}_{E \cdot r} = -\hat{D} \cdot E_0 e^{i \omega t}. \]
(3.12)

By Eq. (3.4), the dipole operator \( \hat{D} \) could be obtained as follows:
\[ \hat{D} = e \sum_{-\frac{\pi}{2a} \leq \vec{k} \leq \frac{\pi}{2a}} \left( \beta(k) \hat{\psi}_{k,s}^\dagger \sigma_2 \hat{\psi}_{k,s} + i \frac{\partial}{\partial \vec{k}} \hat{\psi}_{k,s}^\dagger \hat{\psi}_{k,s} \right), \]
(3.13)

where
\[ \beta(k) = -\frac{\Delta t_0 a}{\varepsilon^2(k)} + u, \]
(3.14)

is the coefficient related to the interband transition between the conduction and valence bands in a unit cell \( 2a \) and the second term in Eq. (3.13) is related to the intraband transition. \( e \) is the electric charge and \( \sigma \) are the Pauli matrixes. \( u \) is dimerized constant related to the lattice distortion. \( \eta \)

**B. Linear response through \( E \cdot r \)**

For the linear susceptibility, \( \chi_{SSH}^{(1)}(\Omega, \omega_1) \) can be obtained from Eq. (1.4) and Eq. (3.13):
\[ \chi_{SSH}^{(1)}(-\omega_1, \omega_1) = 2 \left[ \frac{i}{\hbar} \right] e^2 \sum_k \int_{-\infty}^{\infty} Tr \left\{ i \frac{\partial}{\partial \vec{k}} \left[ G(k, \omega) i \frac{\partial}{\partial \vec{k}} [G(k, \omega - \omega_1)] \right] 
+ \beta(k) \sigma_2 G(k, \omega) \frac{\partial}{\partial \vec{k}} \left[ G(k, \omega - \omega_1) \right] 
+ i \frac{\partial}{\partial \vec{k}} \left[ \beta(k) G(k, \omega) \sigma_2 G(k, \omega - \omega_1) \right] 
+ \beta(k) \sigma_2 G(k, \omega) \beta(k) \sigma_2 G(k, \omega - \omega_1) \right\} \frac{d\omega}{2\pi}, \]
(3.15)

where the Green function \( G(k, \omega) \) is the fourier transformation of \( G(t - t') \equiv -i \langle \hat{T} \hat{\psi}(t) \hat{\psi}^\dagger(t') \rangle \) as follows:
\[ G(k, \omega) = \frac{\omega + \omega_k \sigma_3}{\omega^2 - \omega_k^2 + i\epsilon}, \]
(3.16)

with \( \omega_k \equiv \epsilon(\vec{k})/\hbar \) and \( \epsilon \equiv 0^+ \).

By Eq. (3.17), we have \( \chi_{SSH}^{(1)}(\omega) \equiv \chi_{SSH}^{(1)}(-\omega, \omega) \):
\[ \chi_{SSH}^{(1)}(\omega) = \frac{e^2 (2t_0 a)}{2\pi \Delta^2} \int_{1}^{\frac{1}{2}} \frac{(1 - \eta \delta x^2)^2 dx}{\left(1 - \Delta^2 x^2\right)\left(1 - \delta x^2\right)x^2(x^2 - 1)} \],
(3.17)

where \( x \equiv \hbar \omega_k/\Delta \), \( z \equiv \hbar \omega/(2\Delta) \), \( \delta \equiv \Delta/(2t_0) \) and relative distortion \( \eta \equiv (2u)/a \). Eq. (3.17) can be numerically integrated if one change \( x \to x + i\epsilon \) in considering the life-time of the state. For polyacetylene, by choosing \( t_0 = 2.5eV, \Delta = 0.9eV, a = 1.22A, u = 0.04A \) and \( \epsilon \sim 0.03 \). \( \Delta = 0.18 \) and \( \eta = 0.07 \). The value of \( |\chi_{SSH}^{(1)}| \) with or without \( \eta \) contribution are plotted in Fig.1. As we can clearly see from the graph, the relative distortion \( \eta \)'s contribution is very small (about 1%). We can see the ‘unklapp enhancement’ peak at \( z = 1 \) comparing with the peak \( z = 1/\delta \). Those results are also discussed in the previous works.
FIG. 1. $|\chi_{SSH}^{(1)}(\omega)|$ with $z \equiv \hbar \omega/(2\Delta)$, for $\epsilon = 0.03$, $\delta = 0.18$ and for $\eta = 0.07$ (solid line) or for $\eta = 0$ (dashed line).

If the continuum limitation is applied, that is, $\delta \to 0^+$, $\eta \to 0^+$, $\epsilon \to 0^+$ and $2\hbar \sigma \to \hbar v_F$, the above integral Eq.(3.17) approaches the linear optical susceptibility $\chi_{TLM}^{(1)}(\omega)$ under the TLM model [25] as follows:

$$\chi_{TLM}^{(1)}(\omega) = -\frac{e^2 h v_F}{2\pi \Delta^2 z^2} (1 - f(z)),$$

where

$$f(z) \equiv \begin{cases} \frac{\arcsin(z)}{z\sqrt{1-z^2}} & (z^2 < 1), \\ -\frac{\cosh^{-1}(z)}{z\sqrt{z^2 - 1}} + \frac{i\pi}{2z\sqrt{z^2 - 1}} & (z^2 > 1). \end{cases}$$

The conductivity $\sigma(\omega)$ given by $-i \omega \cdot \chi^{(1)}$. The $\text{Re} [\sigma^{(1)}(\omega)]$ is the exact same as previous results. [20,23]

The calculated $\chi_{TLM}^{(1)}$ and absolute value of $\chi_{TLM}^{(1)}$ are shown in Fig.2 and Fig.3.

FIG. 2. The real part (solid line) and the imaginary part (dashed line) of $\chi_{TLM}^{(1)}(\omega)$ with $z \equiv \hbar \omega/(2\Delta)$.
The above computations indeed are based on perturbative scheme with $\hat{\psi}_{k,s}^\dagger(t)$ and $\hat{\psi}_{k,s}(t)$ as unperturbed creation and annihilation operators under Hamiltonian $\hat{H}_0$. [Eq. (3.11)] From the above figures and expressions, it is very obvious that the DD correlation approach (or $E \cdot r$) will not have ZFD in $\chi^{(1)}$ and $\sigma^{(1)}$. The straightforward computations easily show that Eq.(3.17) and Eq.(3.18) obey the KK relation. Those results are certainly reasonable under the physical picture.

C. The qualitative different results through the polarization operator $\hat{P}$

If the above dipole operator $\hat{D}$ is replaced by the polarization operator $\hat{P}$ defined by Eq.(3.2), which are extensively used in the models, [7] we will obtain a different result from Eq.(3.18). As pointed in the literatures, [13,16,17,23] the polarization operator $\hat{P}$ is sensitive to boundary conditions and singular in the thermodynamic limit. We will demonstrate this issue based on $\hat{P}$ in SSH model. Although the original point of position $R_l$ can be arbitrary, we can pick up the chain region $l$ from 1 to $N$ units to do a simple test. By Eq.(3.2), Eq.(3.6)-Eq.(3.9), we obtain the unit polarization $\hat{P}^{\text{unit}}(k)$ from the total polarization $\hat{P}^{\text{total}}(k)$ in the momentum space:

$$\hat{P}^{\text{unit}}(k) = \lim_{N \to \infty} \frac{\hat{P}^{\text{total}}(k)}{N} = \frac{e a}{2} \sum_{k,s} \psi_{k,s}^\dagger \sigma_2 \psi_{k,s}$$

We find that $\hat{P}^{\text{unit}}(k)$ contains no intraband transition like 2nd term in Eq.(3.13). Applying a similar computation as DD correlation, we can obtain linear susceptibility $\chi^{(1)}_{SSH_P}$ based on $\hat{P}^{\text{unit}}(k)$:

$$\chi^{(1)}_{SSH_P}(\omega) = \frac{e^2 (2t_0 a)}{2\pi (2t_0)^2} \left\{ \int_1^{\frac{\pi}{2}} \frac{dx}{[(1-\delta^2 x^2)(x^2-1)]^{\frac{3}{2}}} + \int_1^{\frac{\pi}{2}} \frac{z^2 dx}{[(1-\delta^2 x^2)(x^2-1)]^{\frac{3}{2}}(x^2-z^2)} \right\}$$

$$= \frac{e^2 (2t_0 a)\delta^2}{2\pi \Delta^2} \left\{ F(\frac{\pi}{2}, \sqrt{1-\delta^2}) + \int_1^{\frac{\pi}{2}} \frac{z^2 dx}{[(1-\delta^2 x^2)(x^2-1)]^{\frac{3}{2}}(x^2-z^2)} \right\},$$

(3.21)

where $F(\pi/2, \sqrt{1-\delta^2})$ is the complete elliptic integrals of the first kind. Obviously, the 1st term in the above Eq.(3.21) is a constant and it is logarithm singular if $\delta \to 0^+$. This result obeys the previous conclusions obtained in solid states, [13,16,17,23] showing that the polarization operator’s failure in optical susceptibility’s study. We choose the same parameter as in Sec.III.B to plot spectrum of $|\chi^{(1)}_{SSH_P}|$, the comparison curves plotted in Fig.4 showing that the results computed from $\hat{D}$ and $\hat{P}$ are actually different both in the spectrum and in the magnitude. We find another peak at $z = 1/\delta$ corresponding to the transition from the bottom of valence band to the top of conduction band is greater than the peak at $z = 1$. The ‘unklapp enhancement’ disappears although the same position of resonant peak still can be predictable based on $\hat{P}$. Those differences show that the electron transition from all the other sites can not be neglectable especially in the periodic models. And conclusions obtained in $\chi^{(1)}$ on the dependence of gap parameter...
\[ \Delta \text{ are qualitatively different for an infinity chain between } \hat{D} \text{ and } \hat{P}. \] Therefore there are some nonconsistencies between the polarization operator \( \hat{P} \) and the dipole operator \( \hat{D} \) at least in the linear response theory.

**IV. LINEAR RESPONSE THROUGH CURRENT-CURRENT CORRELATION**

**A. SSH Hamiltonian in the vector potential form**

The tight-binding Hamiltonian under \( \mathbf{p} \cdot \mathbf{A} \) form should be invariant under gauge transformation (Eq.(2.2) and Eq.(2.3)). \[ \overline{23,26} \] If we change the phase of the one-particle wave function in tight-binding approximation:

\[ \tilde{C}_s'(\mathbf{r}) = e^{i\theta} \tilde{C}_s(\mathbf{r}). \tag{4.1} \]

we must modify the kinetic energy term according to the unperturbed Hamiltonian \( \hat{H}_0 \) in the Wannier functions basis as follows:

\[ H_0 \equiv \sum_{s, \mathbf{r}, \mathbf{r}'} t(\mathbf{r} - \mathbf{r}') (C_{s}(\mathbf{r}) + C_{s}^\dagger(\mathbf{r}'))\tilde{C}_s'(\mathbf{r}) + \text{h.c.} \] \tag{4.2}

\[ \rightarrow \sum_{s, \mathbf{r}, \mathbf{r}'} t(\mathbf{r} - \mathbf{r}') \left[ C_{s}^\dagger(\mathbf{r}) e^{-i\int_{\mathbf{r}}^{\mathbf{r}'} d\mathbf{x} \mathbf{A}(\mathbf{x})/\hbar} C_{s}^\dagger(\mathbf{r}') 
+ C_{s}^\dagger(\mathbf{r}') e^{i\int_{\mathbf{r}}^{\mathbf{r}'} d\mathbf{x} \mathbf{A}(\mathbf{x})/\hbar} C_{s}(\mathbf{r}) \right], \tag{4.3} \]

where \( t(\mathbf{r} - \mathbf{r}') \) is the hopping from position \( \mathbf{r} \) to \( \mathbf{r}' \). \( C_{s}^\dagger(\mathbf{r}) \) creates an electron at site \( \mathbf{r} \) with spin \( s \), \( q \) is the particle charge. The above transformation is also known as Peierls substitution. \[ \overline{23} \] The above form Eq.(4.3) has some kind of general meanings in TBA and can be frequently seen in the theoretical works. \[ \overline{12} \]

If the function \( f(\mathbf{r}, t) \) is arbitrary in Eq.(2.2), it is easy to verify that the above TBA Hamiltonian \( \hat{H}_0 \) is an invariant if the local phase change according to Eq.(4.1) or Eq.(2.3):

\[ \theta(\mathbf{r}, t) \equiv \frac{e}{\hbar} f(\mathbf{r}, t). \tag{4.4} \]

As a specific example, the SSH Hamiltonian with the vector potential \( A \) should be as follows (we change \( \mathbf{A} \) into \( A \) since it is 1d case): \[ \overline{43} \]

\[ H_{SSH}(A) = -\sum_{l,s} \left[ t_0 + (-1)^l \frac{\Delta}{2} \right] (\tilde{C}_{l+1,s}^\dagger e^{-ieA(R_l-R_{l+1})/\hbar} \tilde{C}_{l,s} + \tilde{C}_{l,s}^\dagger e^{ieA(R_l-R_{l+1})/\hbar} \tilde{C}_{l+1,s}), \tag{4.5} \]
B. result without gauge phase factor

Under the assumption of the gauge phase factor (4.4) as no physical meanings, [3] we ignore the phase θ and treat the creation and annihilation operators \{\hat{C}^\dagger\} and \{\hat{C}\} the same as unperturbed creation and annihilation operators \{\hat{C}^\dagger\} and \{\hat{C}\} defined in Eq.(3.1).

The above A Hamiltonian Eq.(4.4) can be expanded in the powers of the external vector potential A and obtain the following:

\[ \hat{H}_{SSH}(A) = \hat{H}_0 - \hat{J}_0 A - \frac{1}{2} \hat{J}_1 A^2 + O(A^3), \quad (4.6) \]

where \( H_0 \) is given by Eq.(3.1),

\[ \hat{J}_0 = - \sum_{l,s} \frac{e}{\hbar} \left[ t_0 + (-1)^l \frac{\Delta}{2} \right] \left[ a - 2(-1)^l u \right] \]
\[ \left( \hat{C}^\dagger_{l+1,s} \hat{C}_{l,s} - \hat{C}^\dagger_{l,s} \hat{C}_{l+1,s} \right) \quad (4.7) \]

and

\[ \hat{J}_1 = - \sum_{l,s} \frac{e}{\hbar} \left[ t_0 + (-1)^l \frac{\Delta}{2} \right] \left[ a - 2(-1)^l u \right]^2 \]
\[ \left( \hat{C}^\dagger_{l+1,s} \hat{C}_{l,s} + \hat{C}^\dagger_{l,s} \hat{C}_{l+1,s} \right), \quad (4.8) \]

The current operator \( \hat{J} \) is obtained from the following equation:

\[ \hat{J} = \frac{i}{\hbar} \left[ \hat{P}, \hat{H} \right]. \quad (4.9) \]

From Eq.(3.2) and Eq.(4.6), we obtain the current operator under the SSH Hamiltonian as follows:

\[ \hat{J}_{SSH} = \hat{J}_0 + \hat{J}_1 A, \quad (4.10) \]

where \( \hat{J}_0 \) and \( \hat{J}_1 \) are defined by Eq.(4.7) and Eq.(4.8).

Similar to the computations in Sec.III, we transform the Hamiltonian Eq.(4.6) and the current operators Eq.(4.7) and Eq.(4.8) into the momentum space. By applying Eq.(3.2) and Eq.(3.7), we can obtain the following:

\[ \hat{J}_0(k) = \frac{ea}{\hbar} \sum_{k,s} \left[ A_0(k) \hat{\psi}^\dagger_{k,s}(t) \sigma_3 \hat{\psi}_{k,s}(t) + B_0(k) \hat{\psi}^\dagger_{k,s}(t) \sigma_1 \hat{\psi}_{k,s}(t) \right] \quad (4.11) \]

and

\[ \hat{J}_1(k) = \frac{ea^2}{\hbar} \sum_{k,s} \left[ A_1(k) \hat{\psi}^\dagger_{k,s}(t) \sigma_3 \hat{\psi}_{k,s}(t) + B_1(k) \hat{\psi}^\dagger_{k,s}(t) \sigma_1 \hat{\psi}_{k,s}(t) \right] \quad (4.12) \]

with \( A_0(k), B_0(k), A_1(k) \)and \( B_1(k) \) defined as follows:

\[
\begin{align*}
A_0(k) &= - \frac{(2t_0)^2(1 - \delta^2)\sin(2ka)}{2\varepsilon(k)}, \\
B_0(k) &= - \frac{2t_0\Delta}{\varepsilon(k)} + \eta\varepsilon(k)
\end{align*}
\quad (4.13)
\]

and

\[
\begin{align*}
A_1(k) &= (1 + \eta^2)\varepsilon(k) - \frac{4t_0\Delta\eta}{\varepsilon(k)}, \\
B_1(k) &= \frac{(2t_0)^2\eta(1 - \delta^2)\sin(2ka)}{\varepsilon(k)}
\end{align*}
\quad (4.14)
\]
Following:

Term Eq.(4.19) is similar to the first term in Eq.(3.21) and is a constant independent of analytical form Eq.(4.21) and the Figures, we find that the first term in Eq.(3.18) disappears in this $x$ however, this important feature has not been reported in the previous work. [20,23]

By Eq.(4.15), Eq.(4.18) and Eq.(4.19), we obtain:

$$\chi'_{SSH}^{(1)}(-\omega_1,\omega_1) = \frac{\chi'_{j_0j_0}^{(1)}(-\omega_1,\omega_1)}{-i\omega_1^2} + \frac{\chi'_{J_0}^{(1)}(-\omega_1,\omega_1)}{-i\omega_2^2}, \quad (4.15)$$

with

$$\chi'_{j_0j_0}^{(1)}(-\omega_1,\omega_1) = 2 \left[ \frac{ea}{\hbar} \right]^2 \sum_k \int_{-\infty}^{\infty} Tr \left\{ A_0(k)\sigma_3 G(k,\omega) A_0(k)\sigma_3 G(k,\omega - \omega_1) \right. $$

$$+ A_0(k)\sigma_3 G(k,\omega) B_0(k)\sigma_1 G(k,\omega - \omega_1) $$

$$+ B_0(k)\sigma_1 G(k,\omega) A_0(k)\sigma_3 G(k,\omega - \omega_1) $$

$$+ B_0(k)\sigma_1 G(k,\omega) B_0(k)\sigma_1 G(k,\omega - \omega_1) \left\} \frac{d\omega}{2\pi}, \quad (4.16)$$

$$\chi'_{J_0}^{(1)}(-\omega_1,\omega_1) = -2 i \left[ \frac{ea}{\hbar} \right]^2 \sum_k \int_{-\infty}^{\infty} Tr \left\{ A_1(k)\sigma_3 G(k,\omega - \omega_1) $$

$$+ B_1(k)\sigma_1 G(k,\omega - \omega_1) \right\} \frac{d\omega}{2\pi}, \quad (4.17)$$

where Green function $G(k,\omega)$ is defined by Eq.(4.16). Following the straightforward computations, we can obtain the following:

$$\chi_{j_0j_0}^{(1)}(\omega) = -2 i \frac{e^2(2\hbar a)}{\pi\hbar^2} \int_1^{\frac{1}{2}} \frac{(1 - \eta\delta x^2)^2 dx}{[(1 - \delta^2 x^2)(x^2 - 1)]} \frac{1}{(x^2 - z^2)}, \quad (4.18)$$

and

$$\chi_{J_0}^{(1)}(\omega) = 2 i \frac{e^2(2\hbar a)}{\pi\hbar^2} \int_1^{\frac{1}{2}} \frac{[1 + \eta^2\delta^2 x^2 - 2\eta\delta] dx}{[(1 - \delta^2 x^2)(x^2 - 1)]} \frac{1}{2}, \quad (4.19)$$

Eq.(4.19) is similar to the first term in Eq.(3.21) and is a constant independent of $z$. Eq.(4.19) obviously is not the term $\langle j_0, j_0 \rangle(\omega = 0)$ suggested by Batistic and Bishop. [22]

By Eq.(4.17), Eq.(4.18) and Eq.(4.19), we obtain:

$$\chi_{SSH}^{(1)}(\omega) = \frac{e^2(2\hbar a)}{2\pi \Delta^2 z^2} \left\{ \int_1^{\frac{1}{2}} \frac{(1 - \eta\delta x^2)^2 dx}{[(1 - \delta^2 x^2)(x^2 - 1)]} \frac{1}{2} - \int_1^{\frac{1}{2}} \frac{(1 + \eta^2\delta^2 x^2 - 2\eta\delta] dx}{[(1 - \delta^2 x^2)(x^2 - 1)]} \frac{1}{2} \right\}, \quad (4.20)$$

where $x, \Delta, z, \delta$ and $\eta$ are all the same defined as in Eq.(3.17).

If the continuum limit is applied, we will find that the contribution (Eq.(4.19)) from the diagnostig term ($J_1$ term) disappears. We will obtain the following susceptibility under TLM models:

$$\chi_{TLM}^{(1)}(\omega) = \frac{e^2\hbar_{\chi F}}{2\pi \Delta^2 z^2} f(z), \quad (4.21)$$

where $f(z)$ is defined by Eq.(3.19).

We plot $\chi_{SSH}^{(1)}$, $\chi_{TLM}^{(1)}$, and $|\chi_{TLM}^{(1)}|$ in Fig.5, Fig.6 and Fig.7 with the same parameter as in Sec.III. From the analytical form Eq.(4.21) and the Figures, we find that the first term in Eq.(4.18) disappears in this $J_0J_0$ formula, however, this important feature has not been reported in the previous work. [20,23]
FIG. 5. $|\chi_{SSH}^{(1)}(\omega)|$ through $J_0 J_0$ correlation (solid line) versus $|\chi_{SSH}^{(1)}(\omega)|$ through $DD$ correlation (dashed line) with $z \equiv \hbar \omega/(2\Delta)$, for $\epsilon = 0.03$, $\eta = 0.07$ and $\delta = 0.18$. $|\chi_{SSH}^{(1)}(\omega)|$ obviously shows the ZFD if the gauge phase factor is not considered.

FIG. 6. The real part (solid line) and the imaginary part (dashed line) of $\chi_{TLM}^{(1)}(\omega)$ with $z \equiv \hbar \omega/(2\Delta)$. It shows the ZFD in real part when the gauge phase factor is not considered.

FIG. 7. $|\chi_{TLM}^{(1)}(\omega)|$ through $J_0 J_0$ correlation (solid line) compared with $|\chi_{TLM}^{(1)}(\omega)|$ through $DD$ correlation or $JJ$ correlation through gauge phase factor (dashed line) with $z \equiv \hbar \omega/(2\Delta)$. $|\chi_{TLM}^{(1)}(\omega)|$ shows ZFD.

Under TLM models, the missing diagmagnetic term can be understood directly form the Lagrangian. [44] Obviously, Eq.(4.20) and Eq.(4.21) strongly diverge when $z \rightarrow 0$ for the real part in both Eq.(4.20) and Eq.(4.21). The above results are certainly wrong since it does not follow the KK relation besides this ZFD problem. Careful comparisons
are made between Eq.(4.20), Eq.(4.21), and Eq.(4.17), Eq.(3.18), showing that the correct imaginary parts obtained from the $J_0J_0$ correlation Eq.(4.22) and Eq.(4.3) are still maintained. It also shows that based on $J_0J_0$ correlation, the absorption part (related to imaginary part) will still be correct. This conclusion is not novel to the peoples working on the transport problem, where the Kubo formula based on $J_0J_0$ correlation is already applied as a common knowledge.

Peoples usually resort KK relation to avoid real part ZFD difficulty.\[3\] But how to understand those difficulties (such as ZFD and violation of KK relation in Eq.(4.20) and Eq.(4.21)) becomes a task. The long wavelength approximation used in the above examples already eliminate the possible ZFD caused by the limitation sequence between $k$ and $\omega$.\[12\] Someone maybe argue to solve the ZFD problem by including a diamagnetic term, such as the effective mass $m^*$ and electron density $n_0$ assumption be made in this solid state problem.\[13\] In fact, those assumptions are awkward because the parameters $m^*$ and $n_0$ in diagnostic term cannot be predictable in the models. For example, in the above SSH and TLM models, $m^*$ and $n_0$ could be arbitrary.\[11\] In the TLM model, you cannot include diagnostic term directly from Lagrangian.\[14\] For the SSH model, the diamagnetic term can be included in Eq.(4.10) from the TBA Hamiltonian Eq.(4.5), but you still cannot solve this ZFD problem, as we clearly see from Fig.5 and Eq.(4.23). Moreover, based on diagnostic terms to cancel ZFD, the assumption on the property of medium should be made in the proof.\[14\]

C. solving ZFD by gauge phase factor

As we demonstrate above, the diagnostic term directly obtained from the Eq.(4.5) can not solve the ZFD problem. This ZFD problem, which is a conceptual problem as we have already pointed out in the Sec.II, is caused by the conventional careless treatment of the gauge phase factor in the optical response theory. In this part, we will demonstrate the same result of SSH and TLM models as $DD$ correlation after considering the contribution of gauge phase factor in $JJ$ correlation.

As we discussed in Sec.II, the new creation operator $\hat{C}_l^\dagger$ and annihilation operator $\hat{C}_l$ in Eq.(4.3), should be differed by the gauge phase factor from the unperturbed creation operator $\hat{C}_l^\dagger$ and annihilation operators $\hat{C}_l$. Following Eq.(2.3), we obtain the following after the local phase factor is considered: \[46\]

$$\hat{C}_l = e^{i eA_l / \hbar} \hat{C}_l$$ \hspace{1cm}(4.22)$$

Thus by the above relation, the Hamiltonian in momentum space should make the following change:

$$\hat{H}(k) \rightarrow \hat{H}(\kappa),$$ \hspace{1cm}(4.23)$$

where

$$\kappa = k + \frac{eA}{\hbar}.$$ \hspace{1cm}(4.24)$$

For the SSH Hamiltonian, we have the following new Hamiltonian:

$$H^{new}(k) = H_0^{new}(k) + H_1^{new}(k)A + O(A^2),$$ \hspace{1cm}(4.25)$$

where $H_0^{new}(k)$ defined as the Eq.(3.11) and

$$H_1^{new}(k) = -\frac{ea}{\hbar} \sum_k B_0(k) \psi^{\dagger}_{k,s}(t) \sigma_1 \psi_{k,s}(t).$$ \hspace{1cm}(4.26)$$

The new current operator $\hat{j}^{new}(k)$ could be obtained from the commutator equation $[\hat{D}(k), \hat{H}^{new}(k)]/(i\hbar)$ as the following:

$$\hat{j}^{new}(k) = \hat{j}_{0}^{new}(k) + \hat{j}_{1}^{new}(k)A + O(A^2),$$ \hspace{1cm}(4.27)$$

where $\hat{j}_{0}^{new}(k)$ is exact the same as Eq.(4.11) and

13
\[ \hat{J}_{1}^{\text{new}}(k) = \left( \frac{e a}{\hbar} \right)^2 \sum_{k,s} \left[ A_{1}^{\text{new}}(k)\hat{\psi}_{k,s}^{\dagger}(t)\sigma_{3}\hat{\psi}_{k,s}(t) + B_{1}^{\text{new}}(k)\hat{\psi}_{k,s}^{\dagger}(t)\sigma_{1}\hat{\psi}_{k,s}(t) \right] \]  

(4.28)

where

\[
\begin{align*}
A_{1}^{\text{new}}(k) &= \frac{B_{0}^{2}(k)}{\varepsilon(k)}, \\
B_{1}^{\text{new}}(k) &= -A_{0}(k) \left[ \frac{2\eta\Delta}{\varepsilon^{2}(k)} + \eta \right].
\end{align*}
\]

(4.29)

where \(A_{0}(k)\) and \(B_{0}(k)\) are defined in Eq.(4.13).

Thus, we find that after considering the gauge factor, the new current operator \(J_{0}^{\text{new}}(k)\) is the same as the static current \(J_{0}(k)\), but \(J_{1}^{\text{new}}(k)\) is different from the static current \(J_{1}(k)\), we call those current differences between \(J_{1}^{\text{new}}(k)\) and static currents \(J_{0}(k), J_{1}(k)\), etc., as induced field currents (IFC) since they are introduced by the gauge field.

Through the evolution operator in interaction picture, \(\ddot{\chi}_{jj}^{(1)}(\omega, \omega_{1}) = \chi_{j_{0}, j_{0}}^{(1)}(-\omega, \omega_{1}) + \chi_{j_{1}, j_{1}}^{(1)}(-\omega, \omega_{1}) - \frac{\omega_{1}^{2}}{\pi} \) as follows:

\[ \chi_{j_{0}, j_{0}}^{(1)}(-\omega, \omega_{1}) = \frac{1}{\hbar} \sum_{k} \int_{-\infty}^{\infty} \text{Tr} \left\{ \right. \\
\quad \left. A_{0}(k)\sigma_{3}G(k, \omega)B_{0}(k)\sigma_{1}G(k, \omega - \omega_{1}) \right. \\
\quad + B_{0}(k)\sigma_{1}G(k, \omega)B_{0}(k)\sigma_{1}G(k, \omega - \omega_{1}) \left. \right\} \frac{d\omega}{2\pi}. 
\]

(4.31)

While the contribution from \(J_{1}^{\text{new}}\) in Eq.(4.32) can be obtained as:

\[ \chi_{j_{1}, j_{1}}^{(1)}(-\omega, \omega_{1}) = -2\left( \frac{e a}{\hbar} \right)^{2} \sum_{k} \int_{-\infty}^{\infty} \text{Tr} \left\{ \right. \\
\quad \left. A_{1}^{\text{new}}(k)\sigma_{3}G(k, \omega - \omega_{1}) \right. \\
\quad + B_{1}^{\text{new}}(k)\sigma_{1}G(k, \omega - \omega_{1}) \left. \right\} \frac{d\omega}{2\pi}. 
\]

(4.32)

Eq.(4.31) gives exact the same result as Eq.(4.18), which is computed through \(J_{0}J_{0}\) correlation. While the contribution from \(J_{1}^{\text{new}}\) in Eq.(4.32) can be obtained as:

\[ \chi_{j_{1}, j_{1}}^{(1)}(\omega) = \frac{2i}{\pi\hbar^{2}} \int_{1}^{\frac{\hbar}{\pi}} \frac{(1 - \eta x^{2})^{2} dx}{[(1 - \delta^{2}x^{2})(x^{2} - 1)]^{1/2} x^{2}}. 
\]

(4.33)

The above term \(\chi_{j_{1}, j_{1}}^{(1)}(\omega)\) actually is \(\langle [j_{0}, j_{0}] \rangle (\omega = 0)\) suggested by Batistic and Bishop. \(22\)

By Eq.(4.18) and Eq.(4.33), Eq.(4.30) returns the exact the same result as Eq.(3.14) and Eq.(3.18), which are computed through \(DD\) correlation.

V. DISCUSSIONS

A. equivalence condition between two gauges

The above examples based on \(J_{0}J_{0}\) and \(DD\) correlations show the importance of the gauge phase factor \(F_{g}\) in the optical response theory. The equivalence of the two gauges \((\mathbf{E} \cdot \mathbf{r} \cdot \mathbf{p} \cdot \mathbf{A})\) should not be based on the exact same sets of wave functions, but should be different by the gauge phase factor. This crucial point has never been clearly pointed out previously. The conventional equivalence between the static current-current \((J_{0}J_{0})\) correlation and the static dipole-dipole \((DD)\) correlation is not maintained, instead, we should consider the induced field currents (IFCs) which is introduced by the gauge phase factor. For example, in the periodic models, we should do the phase shift from \(k \rightarrow \kappa\) for the basis wave functions (or through creation and annihilation operators in models) to obtain the new current operator \(J_{1}^{\text{new}}\) for linear case. Thus, the equivalence between current-current and dipole-dipole correlations should be understood as the situation when all IFCs are included. This paper’s conclusion that the time-dependent gauge phase factor \(F_{g}\) can not be ignored under the perturbative scheme is also consistent with Langhoff et al.'s results. \(40\)
B. Initial distribution function in two gauges

In optical response, people are much more interested in the population of the states than the phase factor. Recently, femtochemistry experiments are able to reveal the phase factor’s effect by the vibrational modes of nuclei other than the population effect (or distribution function) of states. To some extend, the phase factor’s influence on the optical response begin to raise people’s interest. Although our treatment of the optical susceptibilities is still under the frame of non-vibration nuclei, the gauge phase factor’s effect in the theory still can be seen from the above computations.

Another important feature caused by gauge phase factor $F_g$ is the influence on the initial distribution function $f_n(k)$ between two gauges. If all particles are in the ground state $\psi^0_g$ of unperturbed Hamiltonian $\hat{H}_0$ when the electric field $E$ is applied at time $t = 0$, under the perturbative scheme, we can use the set of unperturbed wave functions $\{\psi^0_n\}$ with the initial distribution function

$$f_g(k) = 1 \quad \text{and} \quad f_n(k) = 0 \quad \text{for all the other}(n \neq g); \quad (5.1)$$

Previously, we use the exact the same initial distribution function such like Eq.(5.1) for both gauges.

As we show in the previous sections, the exact wave functions between two gauges should be different by a time-dependent gauge phase factor. Those conclusions are also kept under the perturbative scheme. Thus, the initial distribution function in two different gauges are not necessary the exact same as Eq.(5.1) and should be carefully considered on the choice of the basis sets.

Specifically for $E \cdot r$, we should choose unperturbed wave functions as the basis to avoid ZFD directly. Then the initial distribution function could be Eq.(5.1), since the initial ground state is $\psi^0_g$. But for $p \cdot A$, since the new ground state should be $e^{iF_s} \psi^0_g$ according to Eq.(2.3), there are two ways to set the initial distribution function: (i)If we have already considered the gauge phase factor in our new basis set, that is, we use $\{e^{iF_s} \psi^0_n\}$ as our basis, the distribution function is still as Eq.(5.1); (ii) If we still use unperturbed wave function $\{\psi^0_n\}$ as our basis, we should project the initial wave function $e^{iF_s} \psi^0_g$ on the basis set $\{\psi^0_n\}$ instead of directly applying Eq.(5.1) as our initial distribution function. The latter way is much more complicated since the initial set of distribution function will be time-dependent. In the previous example we show, we use the first way under the local phase approximation. Both ways to treat initial distribution function by $p \cdot A$ are complicated, in this meaning, the gauge $E \cdot r$ is much better. This conclusion has actually already been tested in the practical applications.

C. some previous puzzles

In understanding those nonequivalent results between the two gauges, some literatures are emphasized by the others. However, those explanations lacks the direct proof and the effect of the gauge phase factor is ignored. The above two subsections provide the qualitative reasons to understand those nonequivalent puzzles between the two gauges in the numerical computations. Based on the same set of unperturbed wave functions, the computations of Bassani et al. shows that the $E \cdot r$ converges much faster than $p \cdot A$ reveal that the distribution Eq.(5.1) is good for $E \cdot r$ but not for $p \cdot A$ case, which is certainly reasonable according to our discussions in Sec.V.B. The $\chi^{(2)}$ computation is a special case, since some terms could be 0 if the symmetry is applied. Unfortunately, the nonequivalent results between $J_0J_0$ correlation and $DD$ correlation are magnified in the $\chi^{(3)}$ computations. As an example, the spectrum of $\chi^{(3)}$ of polyacetylene shows a different spectrum between two gauges. Those difference could be understood qualitatively through the gauge phase factor.

VI. CONCLUSIONS

From the computations and discussions above, we concluded that Eq.(1.2) based on the static current is improper and leads ZFD problem, thus, IFCs generated from the gauge phase factor should be included to solve this difficulty. Generally speaking, the Hamiltonians under two gauges are not necessary equivalent unless the gauge phase factor is properly considered through the wave functions. Because the choice problems in the initial distribution function and the basis sets under two gauges (Sec.V.B), it will be very tedious to do the perturbative computations based on the $p \cdot A$ than $E \cdot r$. If the careful computation is made based on the concept of gauge phase factor, both gauges will lead to the equivalent result. Although our computations are chiefly based on a 1d periodic model, it could be easily seen that the chief conclusions of this paper can be expanded in the 2d, 3d and other systems based on the general illustrations in Sec.II.
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does the following $\hat{D}\&\hat{J}$ replacement: $\hat{D} \rightarrow \hat{J}/(i\omega)$. (See p.107. [3]) Although in Bloembergen’s notes, [5] the $A^2(t)$ is seemingly included in $\chi^{(2)}$ derivations(p.35), the results obtained by Bloembergen (Eq.(2-48)) are exactly the same as the results without considering the $A^2(t)$ terms. (See Eq.(4.61) by Butcher and Cotters [3] with $\hat{D}\&\hat{J}$ replacement.) It shows that $\chi^{(2)}$ computations is a special case in nonlinear susceptibility. [13] And in $\chi^{(3)}$ derivations on p.172, [5] the expansion basis is the unperturbed wave functions.

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[44] The Lagrangian for TLM models is described as $L = \int dx \bar{\psi}(x)\{i\bar{\hbar}\partial_t + i\sigma_3v_F \partial_x + \sigma_1\Delta\}\hat{\psi}(x)$. The vector potential $A$ is included by changing $-i\hbar\partial_x \rightarrow -i\hbar\partial_x - eA$, it does not contain $A^2$ term directly from the Lagrangian.
[45] Based on the polarization $\hat{P}$, the behavior of $\chi^{(1)}$ is dependent of $\Delta^{-1}$ and is qualitatively different from the result under $\hat{D}$, which is commonly considered to be dependent of $\Delta^{-2}$, although it was argued that the optical gap $\tilde{\omega}(\Delta)$ should be introduced in understanding those difference between $\hat{P}$ and $\hat{D}$. Please see [S. Tretiak, V. Chernyak, and S. Mukamel, Phys. Rev. Lett. 77, 4656 (1996)].
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