Non-equilibrium nature of non-linear optical response: Application to the bulk photo voltaic effect

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The bulk photo-voltaic effect is an example of a non-linear optical response that leads to a DC current that is relevant for photo-voltaic applications. In this work, we theoretically study this effect in the presence of electron-phonon interactions. Using the response function formalism we find that the non-linear optical response, in general, contains three operator correlation functions, one of which is not ordered in time. This latter correlator cannot be computed from equilibrium field theory. Using a semiclassical approach instead, we show that the bulk photo-voltaic effect can be attributed to the dipole moment of the generated excitons. We then confirm the validity of the semiclassical result (which agrees with the non-interacting result) for non-linear DC response from a quantum master equation approach. From this formalism we find that, in contrast to usual linear response, the scattering rate has a strong implicit effect on the non-linear DC response. Most interestingly, the semiclassical treatment shows that the non-linear DC response for spatially inhomogeneous excitation profiles is strongly non-local and must involve out-of-time-ordered correlators that cannot be computed by equilibrium field theory.

I. INTRODUCTION

The fluctuation-dissipation theorem that relates the perturbative response of observables to equilibrium correlators of observables is one of the corner stones of quantum many-body physics. Recently, a number of non-linear phenomena such as non-linear optical response, photogalvanic effect, pump-probe spectroscopy have been used to characterize solid state materials. For example, certain second harmonic generation coefficients have been used to detect a hidden inversion symmetry breaking nematic phase [1]. Similar correspondence has been shown between second harmonic generation and inversion symmetry breaking in certain topological materials with hexagonal warping term and a gap for quasiparticle excitations[2, 3]. The non-linear response of the DC current in a Weyl material to circularly polarized light, the so-called circular photogalvanic effect has been shown to be quantized [4]. In fact, the quantized circular photogalvanic response has been directly related to the topological monopole charge of Weyl materials [4].

The circular photogalvanic effect is one example of a broader class of bulk photo-voltaic effects where the non-linear response to optical excitation results in a DC current. The bulk photo-voltaic effect (BPVE) for linearly polarized light has been argued to be a result of shift currents [5] in bulk materials, where the current is generated by the sequential tunneling of electrons to one direction. It was also shown that the magnitude of this response is directly related to the Berry curvature over parts of the Brillouin zone where the optical perturbation is resonant [6, 7]. Aside from fundamental interest as a probe of Berry curvature, the BPVE also has potential applications to solar cell devices provided materials with high efficiency can be found [8–16].

Despite the use of characterizing symmetries and topology of materials, the understanding of non-linear optical responses such as the BPVE is quite limited outside the purview of non-interacting systems. As elaborated in Sec. II, the response function formalism for non-linear effects such as the BPVE contains terms that are neither time-ordered nor anti-time ordered. Such out of time ordered correlated terms that have also been shown to appear in non-linear response of superconductors [17] cannot be computed using standard equilibrium Feynman diagram formalism and in this sense are truly non-equilibrium properties.

The utility of the BPVE for solar cell devices ultimately relies on the ability to annihilate photons and convert them to electrical power. However, in the equilibrium diagram formalism, each power of the vector-potential $A(t)$ is associated with either the absorption or emission of a photon. The creation of a DC current at second order in a vector potential would then have to be interpreted as the sequential absorption and emission of a photon. Within this picture, the generation of electrical energy appears quite paradoxical. An electrical current can only generate...
electrical energy in the presence of a finite voltage that can arise in the presence of a scattering process, which is ignored in the standard formalism for shift currents [5–7]. Conceptually, it is desirable to develop a formalism for the BPVE that includes such scattering processes in a way that is completely consistent with energy conservation.

In this work, we systematically study the BVPE in a minimally interacting system, where we include the interaction of the electrons with the phonons in the material. In Sec. II, we start by developing a response function formalism for the BPVE and demonstrating the role of out of time-ordered correlators. The presence of such correlators invalidates the standard computation using equilibrium Feynman diagrams. In order to gain intuition, we first develop a semi-classical picture of the BPVE (Sec. III) based on the processes shown in Fig. 1. This picture is shown to lead to the conventional expression for the DC current in the case of homogeneous optical excitation. However, the application of inhomogeneous excitation profiles are found to lead to results that cannot be described by equilibrium response functions. Such a semiclassical treatment is only justified in the strong electron-phonon scattering limit relative to such correlators invalidates the standard computation using equilibrium Feynman diagrams. Finally, in Sec. V, we study the cross-over between the small and large vector potential, electron-phonon scattering. Finally, in Sec. V, we study the cross-over between the small and large vector potential limit by computing the response numerically within the same formalism for the Rice-Mele model [18].

II. SECOND ORDER OPTICAL RESPONSE

We consider a generic Hamiltonian for a two band semiconductor coupled to electromagnetic (EM) field:

\[ \hat{H}(t) = \hat{H}_0 + A(t)\hat{J}(t) \]  
where \( \hat{H}_0 \) is the two band system Hamiltonian and \( \hat{J}(t) \) is the current operator which couples to the EM vector potential, \( A(t) = A_0 e^{\eta t} (e^{i\Omega t} + e^{-i\Omega t}) \equiv A_0 e^{\eta t} \alpha(t) \) where \( \eta = 0_+ \) is the rate at which the EM field is turned on. We can expand \( \hat{J}(t) \) in powers of \( A \) as: \( \hat{J}(t) \simeq \hat{J}_0 + \hat{A}(t)\hat{J}_1 \). The response \( \langle \hat{J}(t) \rangle \), up to second order in \( A_0 \), can be written as

\[ \langle \hat{J}(t) \rangle \simeq A_0 \chi_{lin}(t) + A_0^2 \chi_n(t) + \chi_{otoc}(t) \]

where \( \chi_{lin} \) is linear response given by

\[ \chi_{lin}(t) = t \int_{-\infty}^{t} dt' e^{i\eta t'} \alpha(t') \langle [\hat{J}_0(t'), \hat{J}_0(t)] \rangle \]  

Among the non-linear response functions, \( \chi_n(t) \) is the part of non-linear response that can be viewed as a linear response of the current to a perturbation \( A_0^2 \hat{J}_1(t) \) and is written as:

\[ \chi_n(t) = t \int_{-\infty}^{t} dt' e^{2\eta t'} \alpha^2(t') \langle [\hat{J}_1(t'), \hat{J}_0(t)] \rangle \]  

The term proportional to \( \chi_n(t) \), while non-linear and involving three current operators, is given by the time-ordered product:

\[ \chi_{otoc}(t) = - \int_{-\infty}^{t} dt_1 dt_2 e^{i\eta(t_1+t_2)} \alpha(t_1)\alpha(t_2) \langle \{ T[\hat{J}_0(t_1), \hat{J}_0(t_2)], \hat{J}_0(t) \} \rangle \]  

Here the current operators are in interaction picture defined as \( \hat{J}_{0,1}(t) = 0 \hat{J}_0 \hat{J}_1(t)e^{-iH_0t} \). (\( n \)) means expectation value w.r.t. the ground state of the system. Since \( \chi_{otoc}(t) \) contains out of time ordered correlator (OTOC), Feynman diagrams cannot be used to evaluate it. One requires Keldysh formalism [17] to compute such correlator in presence of any interaction.

Another way to see that the zero frequency part of \( \langle \hat{J}(t) \rangle \) is likely not computable as a response around equilibrium is to quantize the electromagnetic field in terms of photons (as mentioned in the introduction). Expanding \( A(t) = a^\dagger e^{i\omega t} + h.c. \). Substituting, \( A(t) \) into Eq. 2, we see that \( \langle \hat{J}(t) \rangle_{DC} \propto \langle aa^\dagger \rangle \), which represents absorption and remission of a photon. This suggests that no energy is absorbed, even though the DC current in the presence of a voltage would do work. This would likely represent a non-equilibrium situation suggesting the need for a non-equilibrium theory. Consequences of this somewhat imprecise argument will become clear from the semiclassical treatment of the DC current in the next section.

III. SEMICLASSICAL DESCRIPTION

In order to understand the absorption of energy more clearly, we include an explicit dissipation process through phonon scattering with a semiclassical approach. In this section we treat electron phonon scattering as a probabilistic process along with the quantum mechanical evolution due to external EM field. For simplicity we consider two band Hamiltonian for a semiconductor which is given by

\[ \hat{H}_0 = \sum_k \Delta_k \left( c_k^\dagger c_k - v_k^\dagger v_k \right) \]  

where \( c_k(v_k) \) is conduction (valence) band state operator and \( 2\Delta_k \) is the band gap.
A. Shift current in homogeneous excitation

The incident EM radiation leads to the creation of electron-hole (e-h) pairs. The time dependent amplitude of such a pair can be extracted from the from the EM coupling in Eq. 1 to be

\[ |\delta \Psi_k(t)\rangle_T \simeq A_0 |J_{0,k}^x - \iota J_{0,k}^y| \sin \left( \frac{\Omega t}{2} - \Delta_k t \right) |e\hbar k\rangle_T \]

\[ = A_0 |J_{0,k}^\perp| e^{\iota \omega_k t} \sin \left( \frac{\Omega t}{2} - \Delta_k t \right) |e\hbar k\rangle_T \]

where \(|0\rangle\) is the ground state and \(|e\hbar k\rangle \equiv e^{\iota \omega_k} |0\rangle\) is e-h pair state. \(J_0 \cdot \sigma \equiv J_0 = \partial_t H(t)|_{A=0}\) such that \(\sigma\) is the vector of Pauli matrices in the band basis (i.e. \(\sigma^x|c_k\rangle = |c_k\rangle, \sigma^y|v_k\rangle = -|v_k\rangle\)) and \(|J_{0,k}^\perp| = \sqrt{(J_{0,k}^x)^2 + (J_{0,k}^y)^2}\).

In non-centrosymmetric materials, the resulting e-h pair can carry a dipole moment. So the electric field generated dipole moment evolves in time accordingly as:

\[ P(t) = \sum_k \langle \delta \Psi_k(t) | (r_e - r_h) | \delta \Psi_k(t) \rangle = A_0^2 \frac{2\pi}{\Omega} \int_{BZ} dk \frac{\sin^2 \left( \frac{\Omega t}{2} - \Delta_k t \right)}{(\Omega - 2\Delta_k)^2} \left( \vec{J}_{0,k} \times \vec{J}_{1,k} \right)_z \]

where \(\vec{J}_{1,k} \equiv \partial_t \vec{J}_{0,k}\). In this semiclassical approach the coherent evolution under the electric field is interrupted by phonon scatterings modeled as a Poisson process. The coherent evolution time \(t\) corresponding to the Poisson process is described by an exponential distribution function as

\[ f(t) = \eta e^{-\eta t} \]

Here \(\eta\) is a physical energy scale corresponding to electron phonon scattering strength. Considering the limit of \(\lim_{\eta \to 0} \frac{\eta}{(\Omega - 2\Delta_k)^2} = \pi \delta(\Omega - 2\Delta_k)\) in Eq. 12 we obtain

\[ \lim_{\eta \to 0} J_{shift}^{DC} = A_0^2 \int_{BZ} dk \left( \vec{J}_{0,k} \times \vec{J}_{1,k} \right)_z \delta(\Omega - 2\Delta_k) \]

which matches with the known expression for shift current given in [4].

The semiclassical approach presented above, is valid in the limit of strong e-ph coupling or weak electric field \((A_0|J_{0,k}^\perp| \ll \eta)\) where the scattering processes are instantaneous events with time scale \(\tau \ll \frac{1}{A_0|J_{0,k}^\perp|}\). In Sec. IV we will use a more quantum mechanical approach to study the case of longer scattering times \(\tau\).

Let us now consider in detail the shifting of electrons by phonons following the coherent exciton generation discussed above. Assuming the phonon scattering matrix elements are momentum independent, the momenta and the corresponding velocities of the electrons and holes are uncorrelated. The resulting motion of the electrons and holes are diffusive starting from the average position. The electrons and holes can recombine and emit a phonon. A current is generated if the electron and hole annihilate with a pair that is different from the way they were created. This is the notion of a shift current. However, if the electron and hole of the generated pair recombine with each other before they can diffuse away, for local electron-hole recombination, the recombined excitons do not have any dipole moment. Therefore, in calculating the shift current, the average dipole moment of the exciton, remains unchanged right until the exciton merges into the electron-hole gas.

B. Finite voltage and size effect on BPVE

Finite voltage that may be generated in the presence of spatial variations can have an effect on the electron-hole gas as mentioned at the end of the last subsection. The carrier density of this electron hole plasma, \(n\), is limited since the rate of exciton generation is balanced by recombination rate so that in steady state, \(n^2 \propto A_0^2\). The shift current in the absence of a voltage can only be stable for perfect translation invariance with periodic boundary conditions. Any variations of parameters in the system that would change the shift current, leads to a finite voltage. Let us now consider boundary conditions different from periodic so that a finite voltage \(V\) can be generated over length \(L\) of the material. The voltage \(V\) will lead to a combination of drift and diffusion current among the carriers which is given by

\[ J^d = \frac{ne^2 \tau_d V}{m} \frac{A_0 V}{L} \]

where \(\tau_d\) is the scattering time for the charge carriers. The combined system with a BPVE together with the
drift current can be thought of as a current source \( J^d \) in parallel with a resistance \( R \propto L/(\tau_d A_0) \). The maximum power that can be extracted from this system is

\[
P_{\text{max,BPVE}} = (J^d)^2 R \propto A_0 V/L. \tag{15}
\]

Let us now consider how a finite voltage is naturally generated when the optical excitation is applied to only part of an otherwise homogeneous system. We assume that the intensity of the optical excitation is uniform over a finite region \( R \) in Fig. 2 and vanishes outside. The uniform optical excitation, according to Eq. 13 leads to a constant shift current \( J^\text{shift} \) in this region. At the same time, the shift (and otherwise total) current must vanish in the insulator outside the region \( R \) (see Fig. 2) where the optical excitation is incident. Since the total current must be divergence free in the steady state, the total current must also vanish in the region \( R \). This occurs by a cancellation of the shift current by the diffusion current \( J^d \) in Eq. 14. Of course, the voltage \( V \) required to create \( J^d \) is not externally applied, but rather generated by the imbalance in electron/hole density resulting from the shift current \( J^\text{DC}_{\text{shift}} \).

The drift/diffusion current \( J^d \) required to cancel the shift current \( J^\text{DC}_{\text{shift}} \) is related to the intensity of the optical excitation in a non-local way. We can see this from the fact that \( J^d \) vanishes if the region \( R \) extends over the entire system and the system has periodic boundary conditions. The diffusion current \( J^d \) even in the middle of the region \( R \) depends entirely on conditions outside region \( R \). The equilibrium response terms \( \chi_{n,2} \) and \( \chi_{n,3} \) in Eq. 2 in an insulator are expected to be relatively local in space. This suggests that the non-local response of the drift current \( J^d \) likely arises from \( \chi_{\text{otoc}} \), which cannot be computed from equilibrium correlations and can in principle be non-local.

IV. QUANTUM APPROACH

In this section we go beyond the limit of weak optical excitation strength by using a more quantum mechanical master equation based approach to the photocurrent. The problem of the semiconductor in EM field can be described by Eq. 1 where \( \hat{H}_0 \) is the Bloch Hamiltonian as in Eq. 7. The EM coupled Hamiltonian (Eq. 1) is time periodic with period \( T \equiv 1/\Omega \). The time periodic Hamiltonian in Eq. 1 can be expressed in Floquet basis as

\[
\hat{H}_F = \sum_k \left( \epsilon_k f^\dagger_k f^\mu_k + \epsilon_k^d f^d_k f^\mu_k \right) \tag{16}
\]

where \( \epsilon_k^{u,d} \in [0,\Omega] \) are the two quasi energies and \( f^u,d_k \) correspond to Floquet state operators (see Appendix A for derivation of Floquet spectrum). In the quantum mechanical treatment, we include the phonons explicitly with a Hamiltonian that is written as

\[
\hat{H}_p = \sum_q \omega_q^s a_q^\dagger a_q^s \tag{17}
\]

where \( a_q^s \) is annihilation operator for phonon of mode \( s \) and momentum \( q \). In absence of any e-ph coupling the evolution operator for combined e-ph system is given by

\[
\hat{U}_0 \equiv \hat{U}_F \otimes \hat{U}_p = e^{-i\hat{H}_F t} \otimes e^{-i\hat{H}_p t}. \tag{18}
\]

The phonons couple to the electrons through the e-ph interaction term in the Hamiltonian given by

\[
\hat{V} = \sum_{q=k-g} \left( V_1 c_k^\dagger c_g + V_2 v_k^\dagger v_g \right) \left( a_q^s + a_q^d \right) \tag{19}
\]

where \( V_1 (V_2) \) is the amplitude of intra band scatterings within conduction (valence) band.

The equation of motion for e-ph combined density operator \( \hat{\rho}_I(t) \), in the interaction picture basis that is rotating according to Eq. 18, is given by

\[
\partial_t \hat{\rho}_I = -\frac{i}{\hbar} \left[ \hat{V}_I, \hat{\rho}_I \right]. \tag{20}
\]

Note that \( \hat{V}_I \) is obtained from Eq. 19 by transforming the field operators \( c_k, v_k \) and \( a_q^s \) to the interaction picture basis. In a standard procedure (outlined in Appendix B) to obtain the dynamical equation for the reduced electronic density operator, \( \hat{\rho}_I \), Eq. 20 is solved up to second order in \( |V_1,2| \) following which the phonon operators are traced out under Markovian approximation. The resultant equation of motion for \( \hat{\rho}_I \) becomes

\[
\partial_t \hat{\rho}_I = -Tr \left\{ \hat{V}_I(t), \int_{-\infty}^t dt' \left[ \hat{V}_I(t'), \hat{\rho}_I(t) \right] \right\}_p \tag{21}
\]

\( \hat{\rho}_I \) can be represented more concretely in terms of its matrix elements in Floquet basis:

\[
\Pi_k^{ab}(t) \equiv Tr \left\{ \hat{\rho}_I(t) f^a_k f^b_k \right\}_e \equiv Tr \left\{ \hat{\rho}_I(t) U_F^\dagger \Theta_k^{ab} U_F \right\}_e \tag{22}
\]
where in the last line we have used the cyclic property of trace.

A. Equation for equilibrium distribution

To obtain an equation of motion for $\Pi_k^{ab}$ we differentiate Eq. 22 and use Eq. 21 in the similar way described in [19]. In the process we assume zero temperature for the phonon bath and translational invariance for the electronic system (see Appendix B) to obtain the final form of the equation of motion for $\Pi_k^{ab}$

$$\partial_t \Pi_k = -i[\Pi_k, F_k] - \Pi_k A_k - \Pi_k (\hat{I} - \Pi_k) D_k + D_k (\hat{I} - \Pi_k)$$

(23)

where $F_k$ can be thought of re-normalized Hamiltonian (local in $k$) and is given by,

$$F_k^{ab} = \frac{\epsilon_k^a - \epsilon_k^b}{2} \delta^{ab} + \imath V_{kk'} \sum_g \beta_g \left( \tilde{V}_{gg}^{\rho^\beta} - (\tilde{V}_{gg}^{\rho^\beta})^* \right) \Pi_g^{\rho^\beta}$$

(24)

Here the Fourier components of the matrix elements of $V$ and $\tilde{V}$ are given by,

$$V_{k\beta}^{\alpha \beta}(n) = \sum_m \langle \psi_k^{\alpha}(m+n) | \hat{V}^S | \psi_k^{\beta}(m) \rangle,$$

$$\tilde{V}_{k\beta}^{\alpha \beta}(n) = GV_{k\beta}^{\alpha \beta}(n) \Theta (\epsilon_k - \epsilon_g^\beta + n\Omega),$$

(25)

where $|\psi_k^{\alpha}(n)\rangle$ is the $n$th frequency component of the Floquet state $|\psi_k(t)\rangle$ and $\hat{V}^S = \sum_{p, p'} (V_1 c_p^\dagger e_{p'} + V_2 v_p^\dagger e_{p'})$. $G$ is the constant density of states for the phonon bath and $\Theta(x)$ is the Heaviside step function.

The structure of the tensor $D_k$,

$$D_k^{ab} = \sum_g V_{k\beta}^{\rho^\beta} \Pi_g^{\rho^\beta}$$

(26)

in Eq. 23, can be understood by considering the diagonal limit of $\Pi_k$. In this limit the product $\Pi_k^{\rho^\beta}(1 - \Pi_k^{\alpha \alpha})$ along with the corresponding matrix elements represent the probability of scattering from $|\psi_k^{\alpha}\rangle$ to $|\psi_k^{\beta}\rangle$ state. The $\Theta$ function ensures the initial state has higher quasi energy than the final state thus only allowing processes that dissipate energy to the phonon bath. Similarly the tensor $A_k$ is involved in reverse scattering processes (for instance from $|\psi_k^{\alpha}\rangle$ to $|\psi_k^{\beta}\rangle$ scatters) and is given by,

$$A_k^{ab} = \sum_g V_{k\beta}^{\rho^\beta} (\tilde{V}_{k\beta}^{\rho^\beta})^* (\delta^{ab} - \Pi_g^{\rho^\beta}).$$

(27)

The steady state version of Eq. 23 can be obtained by setting $\partial_t \Pi_k = 0$. The result is a set of non linear algebraic equations involving $\Pi_k^{\rho^\beta}(1 - \Pi_k^{\alpha \alpha})$ terms and thus finding exact, simultaneous solutions for $\{\Pi_k^{\rho^\beta}\}$ is numerically intensive.

We note since the EM amplitude is much smaller than the bandwidth of the electronic energy spectrum $|A_0| \lesssim |\Delta(0) - \Delta(\pi)|$, $\Pi_k^{\rho^\beta}$ differs from its ground state value only in a small region (degeneracy region) of $O[A_0]$ in Brillouin zone. Hence scattering contributions between two degeneracy regions (being smaller by order of $A_0$) can be ignored.

Since $\Pi_k^{\alpha \beta}$ is close to its ground state value for almost all of the Brillouin zone, we can use the ground state values for $\Pi_k^{\alpha \beta}$ in the tensors $A_k^{\alpha \beta}, D_k^{\alpha \beta}, F_k^{\alpha \beta}$ to linearize the steady state equation of motion which can be readily solved analytically. The linearized equation of motion, for the $k$ points inside the degeneracy region, is obtained to be (see Appendix C for details)

$$i[\hat{e}_k, \Pi_k] = -\{M_k, \Pi_k\} + \Lambda_k$$

(28)

where $\hat{e}_k = \frac{1}{2} (\epsilon_k^a - \epsilon_k^b) \delta^{ab}$ is the Hamiltonian in Floquet basis. $M_k$ and $\Lambda_k$ can be deduced from $A_k$ and $D_k$ as defined in Eq. 26-27, to be

$$M_k^{\alpha \beta} \approx q \sum_{n<0} \langle \psi_k^{\alpha}(n) | \hat{S}_c | \psi_k^{\beta}(n) \rangle + p \sum_{n \geq 0} \langle \psi_k^{\alpha}(n) | \hat{S}_v | \psi_k^{\beta}(n) \rangle,$$

$$\Lambda_k^{\alpha \beta} \approx 2q \sum_{n<0} \langle \psi_k^{\alpha}(n) | \hat{S}_c | \psi_k^{\beta}(n) \rangle.$$ 

(29)

Here $p$ and $q$ are effective scattering rates for electrons and holes defined as:

$$p \equiv f_2 |V_1|^2 G$$

and $q \equiv f_2 |V_2|^2 G$

(30)

where $f_2$ is the fraction of the Brillouin zone such that $\Omega > 2\Delta_k$. $\hat{S}_c$ and $\hat{S}_v$ are the projector operators on the valence and conduction band subspaces respectively defined as

$$\hat{S}_c \equiv \sum_k c_k^\dagger c_k, \quad \hat{S}_v \equiv \sum_k v_k^\dagger v_k.$$ 

(31)

The solutions of Eq. 28 can be used to calculate current responses.

B. Expression for current response

The matrix elements of current operator in the basis of Floquet states are

$$C_k^{\alpha \beta} = \langle \psi_k^{\alpha} | \hat{J}(t) | \psi_k^{\beta} \rangle.$$ 

(32)

Solving the linear equations in Eq. 28 and calculating $C_k^{\alpha \beta}$ according to Eq. 32 (Appendix D contains expressions for steady state $\Pi_k^{\rho^\beta}$ and $C_k^{\alpha \beta}$), we obtain expression for the current response. The AC current can be obtained by computing the $\pm \Omega$ frequency components of $C_k^{\rho^\beta}$ and combining with the steady state solution for
\( \Pi_{r,\theta}^{\beta} \) (see Appendix D). The resultant response for the AC current up to leading order in electric field amplitude is

\[
J_k^{DC}(\Omega) = A_0 |J_0^{\perp k}|^2 L_k \left[ 1 - \frac{p + q}{\epsilon_k} + \frac{J^2_\perp(\Omega - 2\Delta_k)}{|J_0^{\perp k}|^2} \right]
\]

with

\[
L_k = \frac{\left( \frac{\Omega}{2} - \Delta_k \right)}{2\epsilon_k} \frac{2q + t_k(p - q)}{(p + q)^2 + (2\epsilon_k)^2}
\]

(33)

where \( t_k \) is given by:

\[
t_k \equiv Tr\{\Pi_k\} = \frac{1 \mp \frac{2(p - q)}{p} \frac{\epsilon_k^2}{(p + q)^2 + (2\epsilon_k)^2}}{1 + \frac{(q - p)^2}{pq} \frac{\epsilon_k^2}{(p + q)^2 + (2\epsilon_k)^2}}.
\]

(34)

Similarly, computing the zero frequency component of \( C_{r,\theta}^{\beta} \), we find the DC current up to second order in electric field magnitude to be

\[
J_{DC}^{\perp shif t}(k) = Tr\{\Pi_k C_k\} = 2A_0^2 \left( \vec{J}_0^{\perp k} \times \vec{J}_1^{\perp k} \right)_z \frac{2q + t_k(p - q)}{(p + q)^2 + (2\epsilon_k)^2}.
\]

The total DC current is obtained integrating Eq. 35 over Brillouin zone as

\[
J_{DC}^{\perp shif t} = \frac{1}{2\pi} \int_{BZ} dk J_{DC}^{\perp}(k).
\]

(36)

If the EM perturbation is the smallest parameter (i.e. \( A_0 |J_0^{\perp k}| << p + q \)), from Eq. 34 \( t_k \approx 1 \) which implies

\[
\lim_{A_0 |J_0^{\perp k}| \to 0} \lim_{p,q \to 0} J_{DC}^{\perp shif t}(k) = 2A_0^2 \left( \vec{J}_0^{\perp k} \times \vec{J}_1^{\perp k} \right)_z \frac{2q + t_k(p - q)}{(p + q)^2 + (2\epsilon_k)^2}.
\]

(37)

Recognizing the total scattering rate as \( \eta = p + q \) and using the limit, \( \lim_{A_0 |J_0^{\perp k}| \to 0} \epsilon_k = \frac{\Omega}{2} - \Delta_k \), we recover the semi classical result obtained in Eq. 13.

\[
\lim_{A_0 |J_0^{\perp k}| \to 0} \lim_{p,q \to 0} J_{DC}^{\perp shif t}(k) = A_0 \left( \vec{J}_0^{\perp k} \times \vec{J}_1^{\perp k} \right)_z (2q + t_k(p - q)) \delta \left( \frac{\Omega}{2} - \Delta_k \right).
\]

(38)

Putting this expression into Eq. 36 it is easy to see that the resultant DC current scales linearly with the electric field in this limit, i.e. \( J_{DC}^{\perp shif t} \propto A_0 \). This result contradicts Kubo formula by producing a linear scaling with the external oscillating EM field for the DC current.

V. NUMERICAL EVALUATION OF THE WEAK DAMPING LIMIT

According to the analytic results in Eq. 37 and Eq. 38, the DC current response scales quadratically at small \( A_0 \) and linearly at large \( A_0 \) (smaller and larger than e-ph scattering strengths). To study how the crossover occurs between the two regimes, we perform the integral in Eq. 36 using the expression for \( J_{DC}^{\perp shif t}(k) \) given by Eq. 35 for the Rice-Mele model [18]. We also numerically study the scaling property of AC current magnitude for the same model. The Rice-Mele model is described by the following tight binding Hamiltonian [18]

\[
\tilde{H}_0 = \sum_n \left( h a_n^\dagger b_n + h^* a_n^\dagger b_{n-1} + h.c. \right) + d (a_n^\dagger a_n - b_n^\dagger b_n)
\]

\[
= \sum_k \Delta_k \left( \epsilon_k^2 \epsilon_k - v_1^2 v_k \right)
\]

(39)

with \( \Delta k \equiv \sqrt{h^2 + h'^2 + 2hh'\cos(k) + d^2} \).

The EM vector potential \( A(t) \) introduces phases to the hopping terms according to Peierls substitution as

\[
a_n^\dagger b_n \to e^{-i\frac{\Delta n}{2}} a_n^\dagger b_n, \quad a_n^\dagger b_{n-1} \to e^{i\frac{\Delta n}{2}} a_n^\dagger b_{n-1}.
\]

(40)

Transforming to Fourier space, the perturbed Hamiltonian, in the atomic \((a_k^\dagger, b_k^\dagger)\) basis, becomes

\[
\tilde{H} = \sum_k \left( a_k^\dagger b_k^\dagger \right) \tilde{H}_k \cdot \vec{\sigma} \left( a_k \ b_k \right)
\]

with

\[
\tilde{H}_k = - \left[ \left( h + h' \right) \cos \frac{k - A}{2}, \left( -h + h' \right) \sin \frac{k - A}{2}, -d \right].
\]

(41)

In terms of system parameters, \((\vec{J}_0^{\perp k} \times \vec{J}_1^{\perp k})_z = -\frac{d}{2\Delta_k} (h'^2 - h^2)\) where \( \vec{J}_0^{\perp k} \equiv \partial_A \vec{H}_k(A)|_{A=0} \) and \( \vec{J}_1^{\perp k} \equiv \partial_A^2 \vec{H}_k(A)|_{A=0} \).

The AC current for this Rice-Mele model is obtained by numerical integration of the expression given by Eq. 33 over the Brillouin zone with fixed values for \( p,q \). The resulting values for \( |J^{AC}|/|J_{DC}^{\perp k}| \) are plotted as a function of \( 2A_0 |J_0^{\perp k}^D|/(p + q) \) on a log scale in Fig. 3 where \( k_D \) is the momentum of the degeneracy point (i.e. \( 2\Delta_{k_D} = \Omega \)). The points on the plot fit to a straight line with slope 1. Thus the AC current response scales linearly in a range of values from \( A_0 |J_0^{\perp k}| << (p + q) \) to \( A_0 |J_0^{\perp k}| >> p + q \). This scaling is as expected from conventional linear response theory. On the other hand same plot for \( |J_{DC}^{\perp shif t}|/|J_{DC}^{\perp k}| \) shows a crossover of scaling with \( A_0 \) as shown in Fig. 4. In the limit of weak electric field, \( \left( \ln |2A_0 |J_0^{\perp k}^D|/(p + q)| < -1 \right) \), the points fit to a straight line of slope 2 whereas in the limit of weak scattering \( \left( \ln |2A_0 |J_0^{\perp k}^D|/(p + q)| > 1 \right) \), the corresponding

A
B
A
B
The equation for the linear fit is
\[ y = -11.65 + x. \]

The linear fit equations are
\[ y = -13.3 + 2x \]
and
\[ y = -12.8 + 1.05x \]
respectively.

Thus the e-ph coupling strength sets an energy scale for the EM field strength for a non-linear to linear crossover of the shift current response.

**VI. CONCLUSION**

We have studied an example of a non-linear response i.e. the BPVE in a system with electron-phonon interactions. Since the response is dominated by resonant transitions, we have limited our treatment to a two band model. As we found in Sec. II, the BPVE contains OTOC terms that cannot be computed by simple diagrams. However, in Sec. III, we find that the BPVE can be understood semiclassically from the dipole moment of generated excitons. The semiclassical limit assumes that the scattering rate is large compared to the driving force. We remedy this in Sec. IV using a master equation approach. From this we find that in the small scattering rate limit the DC current scales linearly with the vector potential instead of quadratically. This signals that this result is beyond the response function formalism in Sec. II. The AC current doesn’t show any such anomalous scaling behavior.

The DC current we obtain in the small \( A_0 \) limit is consistent with the non-interacting result for BPVE obtained previously. The breakdown seen in Sec. IV at larger \( A_0 \) shows that electron-phonon interaction affects the result in an implicit way. However, what is interesting from comparing to the general response in Sec. II is that only the \( \chi_{n,2} \) term contribute to the shift current in configurations with homogeneous excitation profiles. This term is really conventional linear response of \( \hat{J}_0 \) to \( \hat{J}_1 \) contributions in the BPVE and can be computed from equilibrium field theory. However, as is clear from the arguments in Sec. III B, the situation is quite different for inhomogeneous excitation profiles. In this case, the excited carrier density \( n \) generated by the incident light can also contribute a current in the case where there are spatial variations of this density. This contribution to the dc current is not captured by the simple shift current that can be computed from \( \chi_{n,2} \) and requires the computation of \( \chi_{otoc} \). As discussed in Sec. II, since \( \chi_{otoc} \) is not a time-ordered correlator, it cannot be computed from conventional equilibrium field theory but rather requires a Keldysh technique. In this work, we have circumvented this using a semi-classical approach that is of limited validity in the context of weak interactions. The full quantum mechanical computation of \( \chi_{otoc} \) using Keldysh field theory for strongly interacting systems where the semi-classical estimate would break down, would be an interesting future direction.

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Appendix A  FLOQUET THEORY

For a system with time periodic Hamiltonian of frequency $\Omega$, the solutions for the time dependent Schrodinger equation (TDSE) have the following form

$$|\psi_k^{\alpha}(t)\rangle = e^{-i\epsilon_k^{\alpha}t}\Phi_k^{\alpha}(t) = e^{-i\epsilon_k^{\alpha}t}\sum_n |\Phi_k^{\alpha}(n)\rangle e^{i\Omega t}.$$  \hspace{1cm} (42)

where the quasi energy $\epsilon_k^{\alpha}$ can be chosen to belong in a $[0, \Omega]$ interval and $|\Phi_k^{\alpha}(t)\rangle$ is time periodic with period $\frac{2\pi}{\Omega}$.

Substituting the form of Eq. 42 in TDSE, one obtains the Floquet equations in frequency space as

$$\sum_n (\epsilon_k^{\alpha} - n\Omega) |\Phi_k^{\alpha}(n)\rangle = \sum_m \tilde{H}_m (m-n) |\Phi_k^{\alpha}(m)\rangle.$$  \hspace{1cm} (43)

where $\tilde{H}_m^{m-n}$ corresponds to the $(m-n)\Omega$ frequency component of the Hamiltonian, $\tilde{H}_m$. The eigenvalue solutions for Eq. 43 produce $\epsilon_k^{\alpha}$ and $|\Phi_k^{\alpha}(n)\rangle$.

For the two band case discussed in the main text, the EM perturbation in frequency space can be written as

$$\hat{H}_p = A_0 \sum_n J^x_{\alpha,\beta} |\psi_{k,n}^{\alpha,\beta}\rangle \langle \psi_{k,n+1}^{\alpha,\beta}| + A_0 \left( J^{y}_{\alpha,\beta} - i J^{y}_{\alpha,\beta} \right) |\psi_{k,n}^{\alpha,\beta}\rangle \langle \psi_{k,n+1}^{\alpha,\beta}| + h.c.$$  \hspace{1cm} (44)

where the subscript $n$ correspond to $n\Omega$ frequency component for the $c_k$, $v_k$ operators.

For the unperturbed Hamiltonian (Eq. 7) the quasi energies are simply

$$\epsilon_k^{\alpha}(0) = \frac{\Omega}{2} + \frac{\Omega}{2} - \Delta_k$$  \hspace{1cm} and  \hspace{1cm} $$\epsilon_k^{\alpha}(0) = \frac{\Omega}{2} - \frac{\Omega}{2} - \Delta_k$$  \hspace{1cm} (45)

with the corresponding states as

$$|\psi_k^{\alpha}(0)\rangle = \Theta(\Omega - 2\Delta_k) |c_k(0)\rangle + \Theta(2\Delta_k - \Omega) |v_k(-1)\rangle,$$

$$|\psi_k^{\beta}(0)\rangle = \Theta(\Omega - 2\Delta_k) |v_k(-1)\rangle + \Theta(2\Delta_k - \Omega) |c_k(0)\rangle.$$  \hspace{1cm} (46)

If $\Omega = 2\Delta_k$, the quasi energies are degenerate and hence to obtain the effect of perturbation (Eq. 44) an exact diagonalisation needs to be performed in the vicinity of those degeneracy points.
Up to leading order in $A_0$, the shifts can be obtained by diagonalizing the following matrix given by

$$
\hat{H}_k = \left( \begin{array}{cc}
\Delta_k & A_0(J^x_{0,k} + \epsilon J^y_{0,k}) \\
A_0(J^y_{0,k} + \epsilon J^x_{0,k}) & -\Delta_k
\end{array} \right)
$$

(47)

The corresponding eigenvectors can be used to obtain the Floquet state solutions in time domain to be

$$
|\psi_k^\alpha(t)\rangle = \frac{e^{-i\epsilon_k^\alpha t}}{\sqrt{1 + |\epsilon_k^\alpha|^2}} (|c_k\rangle \mp x_k e^{-i\Omega t}|v_k\rangle),
$$

$$
|\psi_k^\beta(t)\rangle = \frac{e^{-i\epsilon_k^\beta t}}{\sqrt{1 + |\epsilon_k^\beta|^2}} (|c_k\rangle - x_k e^{-i\Omega t}|v_k\rangle)
$$

with $x_k = \frac{\Omega}{2} - \Delta_k + \epsilon_k J^x_{0,k} + \epsilon J^y_{0,k}$

and $\epsilon_{k}^{\alpha,\beta} = \frac{\Omega}{2} \pm \epsilon_k$ with $\epsilon_k = \sqrt{\left(\frac{\Omega}{2} - \Delta_k\right)^2 + \frac{A_0^2}{2}|J^y_{0,k}|^2}$.

(48)

### Appendix B STEPS OBTAINING RATE EQUATION

Expanding the solution of Eq. 20 up to second order in $|V_{1,2}|$ we obtain:

$$
\hat{\rho}_I(t) = -t \int_{-\infty}^{t} dt' \left[ \hat{V}_{I}(t'), \hat{\rho}_I(-\infty) \right]
$$

$$
- \int_{-\infty}^{t} dt' \left[ \hat{V}_{I}(t'), \int_{-\infty}^{t'} dt'' \left[ \hat{V}_{I}(t''), \hat{\rho}_I(t'') \right] \right]
$$

(49)

Since, the first term contains single power of bath operators, its trace can be dropped. In that case we note, $|\hat{\rho}_I(t') - \hat{\rho}_I(t)| \approx O(|V_{1,2}|^2)$ and so up to second order in $|V_{1,2}|$, we can replace $\hat{\rho}_I(t')$ by $\hat{\rho}_I(t')$ in Eq. 49 to obtain a second order perturbative result for $\hat{\rho}_I(t)$ as

$$
\hat{\rho}_I(t) \equiv Tr \left\{ \hat{\rho}_{I}^{\beta}(t) \right\}_p
$$

$$
- Tr \left\{ \int_{-\infty}^{t} dt' \left[ \hat{V}_{I}(t'), \int_{-\infty}^{t'} dt'' \left[ \hat{V}_{I}(t''), \hat{\rho}_I^{\beta}(t'') \right] \right] \right\}_p.
$$

(50)

Differentiating Eq. 50 w.r.t. time one obtains Eq. 21. Using cyclic property of trace, from Eq. 22, $\partial_t \Pi_{k}^{ab}$ produces

$$
\partial_t \Pi_{k}^{ab} = -i Tr \left\{ \left[ \Omega_{ab}, \hat{H}_k \right] \hat{\rho}_s^{\beta}(t) \right\}_e
$$

$$
- Tr \left\{ \left[ \Omega_{ab}, \hat{U}_0 \hat{V}_{I}(t) \hat{U}_0^{-1} \right] \hat{U}_0 \left( \int_{-\infty}^{t} dt' \hat{V}_{I}(t') \right) \hat{U}_0^{-1} \right\} \hat{\rho}_s^{\beta}(t) \right\}_e.
$$

(51)

Noting that $U_F(t) f_k^{\alpha^1} f_g^{\alpha^2} U_F^{-1}(t) = e^{i(e_k^\alpha - e_g^\alpha)t} f_k^{\alpha^1} f_g^{\alpha^2}$ and $U_F(t) a_g^\dagger U_F^{-1}(t) = a_g^\dagger e^{i\omega_q t}$, we obtain

$$
U_0 \hat{V}_I(t) U_0^{-1} = \sum_{k,g} V_{k}^{\kappa\beta}(t) f_k^{\alpha^1} f_g^{\alpha^2} \left( a_g^\dagger + a_q^\dagger \right)
$$

(52)

where the interaction matrix elements $V_{k}^{\kappa\beta}$ between the Floquet states have the form:

$$
V_{k}^{\kappa\beta}(t) = \sum_n \left[ \sum_m \langle \Psi^\kappa_n(m + n) | \hat{V}^S | \Psi^\beta_m(n) \rangle \right] e^{i\Omega t}
$$

(53)

And the integral over time for the interaction gives

$$
\int_{-\infty}^{t} dt' \hat{V}_{I}(t') = \sum_{p,p'} f_p^{\alpha^1} f_p^{\alpha^2} V_{pp'}^{\alpha^1\alpha^2}(n) a_{p'}^\dagger e^{i(e_q^\alpha + \omega_q^\alpha - n\Omega)t} + \sum_{p,p'} f_p^{\alpha^1} f_p^{\alpha^2} V_{pp'}^{\alpha^1\alpha^2}(n) a_{p'}^\dagger e^{i(e_q^\alpha - \omega_q^\alpha - n\Omega)t}
$$

$$
\left( e_{p'}^\alpha + \omega_{p'}^\alpha - n\Omega + \eta \right)
$$

(54)

where $\eta$ is an arbitrarily small imaginary number introduced to regularize the integrals. Considering only the Cauchy principal value one obtains energy conserving delta functions as follows

$$
U_0 \left( \int_{-\infty}^{t} dt' \hat{V}_{I}(t') \right) U_0^{-1}
$$

$$
= \sum_{p,p'} f_p^{\alpha^1} f_p^{\alpha^2} V_{pp'}^{\alpha^1\alpha^2}(n) a_{p'}^\dagger e^{i\Omega t} \delta \left( e_{p'}^\alpha + \omega_{p'}^\alpha - n\Omega \right)
$$

$$
+ \sum_{p,p'} f_p^{\alpha^1} f_p^{\alpha^2} V_{pp'}^{\alpha^1\alpha^2}(n) a_{p'}^\dagger e^{i\Omega t} \delta \left( e_{p'}^\alpha - \omega_{p'}^\alpha - n\Omega \right)\right).
$$

(55)

Going back to Eq. 51, the first commutator in the second term gives

$$
\left[ \Theta_{ab}^{\kappa\beta}, \hat{V}_I(t) \right] = \sum_{g,q} V_{g}^{\kappa\beta}(t) \left( a_g^\dagger + a_q^\dagger \right) \left[ f_k^{\alpha^1} f_g^{\alpha^2} \right]_{p,q}
$$

$$
= \sum_{k,g} V_{k}^{\kappa\beta}(t) f_k^{\alpha^1} f_g^{\alpha^2} \left( a_g^\dagger + a_q^\dagger \right) - \sum_{k,g} V_{k}^{\kappa\beta}(t) f_k^{\alpha^1} f_k^{\alpha^2} \left( a_q^\dagger + a_q^\dagger \right)
$$

(56)

In order to calculate the second commutator we need to trace out the bath operators from the product of $\Theta_{ab}^{\kappa\beta}, \hat{V}_I(t)$, $\int_{-\infty}^{t} dt' \hat{V}_I(t')$. We use Markovian approximation which assumes that the bath’s response time ($\tau_p$) is much shorter than the dynamical time scale due to interaction Hamiltonian, $\tau_p < < \frac{1}{\Omega}$. Consequently, the phonon density matrix remains effectively constant and can be decoupled from the combined density operator as

$$
\hat{\rho}_I(t) \simeq \hat{\rho}_I(t) \otimes \hat{\rho}_F.
$$

(57)

The decoupling implies $(aa^\dagger f^1 f^1 f^1 f) \simeq (aa^\dagger) (f^1 f^1 f^1 f)$. Now for the bath using zero temperature expectation values, i.e., $(a_q^\dagger a_q^\dagger) = \delta_{qq}$ and assuming continuum energy
Similarly all other matrix element combinations \( (V_{ag}^\alpha(n) V_{bg}^\beta(n)) \) for different \( g \) points (\( \Omega > 2 \Delta g \)) can be obtained. The additional \( \Theta \) function puts constraint on the values of \( n \) that goes into the summation formula in the steady state rate equation. Consequently the contributions from \( g : \Omega < 2 \Delta g \) become \( O(A_k^0) \) and thus can be dropped. Incorporating all these, one obtains the \( M_k \) and \( A_k \) matrix elements as in Eq. 28.

### Appendix D. STEADY STATE SOLUTIONS AND CURRENT MATRIX ELEMENTS

Using the expressions for Floquet states (Eq. 48) one can easily calculate the \( M_k \) and \( A_k \) matrix elements in Eq. 29 to find the solution for Eq. 28 in the form, \( \Pi_k \equiv t_k \hat{J} + \hat{R}_k \cdot \hat{\sigma} \), given by

\[
t_k = \frac{1 - \frac{2(p-q)}{p} \frac{A_k^0 |\psi_{g,k}^\alpha|^2}{(p+q)^2 + (2\epsilon_k)^2}}{1 + \frac{(p-q)^2}{pq} \frac{(p+q)^2 + (2\epsilon_k)^2}{2\epsilon_k}}
\]

\[
\hat{R}^\alpha_k = \frac{2q + t_k(p-q)}{p} \frac{\Omega - \Delta_k}{2\epsilon_k}
\]

\[
\hat{R}^\beta_k = \frac{A_0}{2\epsilon_k} \frac{2q + t_k(p-q)}{(p+q)^2 + (2\epsilon_k)^2} \left( J^\beta_{0,k} - \frac{2\epsilon_k}{p+q} J^\gamma_{0,k} \right)
\]

\[
\hat{R}^\gamma_k = \frac{A_0}{2\epsilon_k} \frac{2q + t_k(p-q)}{(p+q)^2 + (2\epsilon_k)^2} \left( J^\gamma_{0,k} + \frac{2\epsilon_k}{p+q} J^\beta_{0,k} \right)
\]

The current operator, up to first order in \( A_0 \), is given by

\[
\hat{J} = \sum_k \left( \epsilon_{k,m}^\dagger v_{k,m}^\dagger \right) \left( \hat{J}_{0,k} \cdot \hat{\sigma} \right) \left( c_{k,m} \right)
\]

\[
+ A_0 \sum_k \left( \epsilon_{k,m}^\dagger \Delta_{k,m}^\dagger \right) \left( \hat{J}_{1,k} \cdot \hat{\sigma} \right) \left( c_{k,m+1} \right)
\]

Using the Floquet state expressions (Eq. 48) along with the current operator definition in frequency space (Eq. 63), one can obtain the zero frequency component for the matrix elements of current operator, \( C_{k,0}^{\alpha\beta} \), in \( \{ \hat{I}, \hat{\sigma} \} \).
basis to be

\[ \tilde{C}_k^\alpha(0) \equiv \frac{1}{2} \left( \tilde{C}_k^{\alpha\beta}(0) + \tilde{C}_k^{\beta\alpha}(0) \right) \]

\[ \simeq \frac{|x_k|}{1 + |x_k|^2} \frac{J_{0,k} \cdot J_{1,k}}{|J_{0,k}|^2} \left( J_{1,k} \cdot |x_k|^2 \left( J_{0,k} \cdot J_{1,k} \right) \right) \]

\[ \tilde{C}_k^\beta(0) \equiv \frac{1}{2} \left( \tilde{C}_k^{\alpha\beta}(0) - \tilde{C}_k^{\beta\alpha}(0) \right) \]

\[ \simeq \frac{1}{1 + |x_k|^2} \left[ J_{0,k} \cdot J_{1,k} \left( |x_k|^2 \right) + |x_k|^2 \left( J_{0,k} \cdot J_{1,k} \right) \right] \]

with \( sr_k \equiv 1 + \frac{(J_{0,k}^x)^2 - (J_{0,k}^y)^2}{|J_{0,k}|^2} \) and \( si_k \equiv \frac{2J_{0,k}^x J_{0,k}^y |x_k|^2}{|J_{0,k}|^2} \).

The ±Ω frequency components of the matrix elements of current operator, \( \tilde{C}_k^{\alpha\beta}(\pm \Omega) \), are given by

\[ \tilde{C}_k^{\alpha\beta}(\Omega) = \frac{A_0}{2\varepsilon_k} \left( |J_{0,k}^x|^2 + J_{1,k}^z(\Omega - 2\Delta_k) \right) = C_k^{\alpha\beta}(\Omega) \]

\[ \tilde{C}_k^{\alpha\beta}(\pm \Omega) = -C_k^{\alpha\beta}(\pm \Omega) \]

\[ \tilde{C}_k^{\alpha\beta}(\Omega) = \frac{J_{0,k}^x - iJ_{0,k}^y}{1 + |x_k|^2} C_k^{\alpha\beta}(\Omega) = \frac{|x_k|^2}{1 + |x_k|^2} \left( J_{0,k}^x - iJ_{0,k}^y \right) \]

\[ \tilde{C}_k^{\alpha\beta}(\Omega) = \left[ C_k^{\alpha\beta}(\Omega) \right]^* \quad C_k^{\alpha\beta}(\Omega) = \left[ C_k^{\alpha\beta}(\Omega) \right]^* \]

(65)