On time-resolved approach for phonon assisted interband transitions

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Abstract. Photoexcited dynamics of electrons and holes in two-band dielectric, with special emphasis on back reaction of phonons are developed by combining the quantum electrodynamics and Baker-Campbell-Hausdorff (BCH) canonical transformation. These methods create an explicit time-domain representation of photoinduced processes and contribute in unifying phonon-assisted description of distribution functions of electron and hole quasiparticles for the description of observable effects of photoinduced processes in dielectrics.

1. Introduction
Determining the properties of electron-hole (e-h) pairs excited at interaction of electromagnetic radiation with matter is of major interest to many branches of solid state physics. Currently, there are no general efficient methods to deal with this challenging problem that involves joint kinetics of e-h pairs, arbitrary dispersion of the constituting electrons and holes, and their interaction with phonons. Early approaches to this problem categorized as time-convolutionless projection operator techniques were introduced in [1] and provide a systematic way to go beyond the Markovian approximation for quantum two-level systems in a bosonic reservoir [2]. However, these approaches are technically complex and inconvenient for systems with band structure [3].

Complementary developments are based on quantum field theory (QFT) [4] addressed to metastable vacuum, quasiparticle representation of electron and positron, and the associated mathematical technique for the dynamic Schwinger effect. Its extension to lattice systems has lead to a fundamentally new QFT approach to the solid state physics. Similarity between the kinetics of quantum field systems with meta-stable vacuum and the e-h kinetics in solid state is that in both cases the vacuum state is not uniquely defined and the quasiparticle representation is introduced by physical reasons. With application to solids, the vacuum state must obey the concepts of band theory, the conventional quantum mechanics, as well as requirements to the Lorentz invariance and special relativity.

To this end, the Lagrangian density, conventionally derived for a free particle, is addressed to the electron–hole pair and the associated field functions are harmonized with individual dispersion relations of these non-interacting quasiparticles [5]. The electromagnetic radiation is considered as spatially uniform within the atomic scale and is presumably switched on at infinite past. The resulting interband transition obeys the principle of least action for e–h birth and annihilation operators constituting a background for their distribution functions and for the kinetics of observables [6]. Obvious benefits of the QFT approach are reduction of conventional restrictions for the parameters of electromagnetic radiation and non-Markovian nature of the short time e-h kinetics.

A topic, still unresolved and essential for more realistic models, is the Coulomb interaction between photoexcited electrons and holes. Formally this interaction is categorized as back-reaction of phonons which must be distinguished from the impact of thermal (boson) bath at finite temperatures. Available option is to implement additional terms to the QFT Hamiltonian as attempted on the basis...
of the Baker-Campbell-Hausdorff (BCH) canonical transformation[7,8]. However, to justify this ad hoc approximation one needs in a connection between conventional and QFT approaches as a disadvantage.

In the present work we continue our previous attempts as based on a self-consistent description of electrons, holes and the associated phonons. Formally, the back reaction of phonons is accounted for in Lagrangian density on equal grounds with electromagnetic radiation and bridges the conventional band theory with QFT. We are limited within the nondissipative approximation and neglect the interaction with thermal bath.

We start from the free field case to sketch the notations and derivation of the associated Lagrange–Hamilton approach (Section 2). Systematic derivation of field functions is reconsidered for arbitrary dispersion of electrons and holes as a background for further extension toward the back-reaction of phonons (Section 3). A survey of BCH in context of Lagrange–Hamilton approach is given in (Section 4). Conclusions are in (Section 5). We also include Appendix A where the harmonization with dispersion relations and the transition to field operator representation is given. Natural units, $\hbar = c = 1$, are used.

2. Model description: the free field

Derivation of the interband transitions kinetics in the context of e-h pair is thoroughly discussed in the literature [5,6] focussed on quadratic dispersion. However, in order to introduce the notation and to emphasize details specific for the back reaction of phonons we reconsider the survey following the free field, electromagnetic radiation, and phonons stages. The description keeps oscillator-like equation, the Lagrange–Hamilton, and principle of least action representations maintained.

The model two-band system is defined by e-h pair fixed to hypersurfaces in energy - quasi-momentum space with electron and hole states in the conduction and valence bands [5] as

$$E_c = \frac{\Lambda}{2} + \varepsilon_c(p), \quad E_v = -\frac{\Lambda}{2} - \varepsilon_v(p),$$

where the static dispersion laws of the kinetic energy $\varepsilon_c(p)$ and $\varepsilon_v(p)$ are either assumed on the phenomenological basis or calculated by methods of nonrelativistic quantum mechanics. The band gap $\Delta$ and the dispersion laws assigned to conduction and valence states $\varepsilon_{c,v}(p)$ remain presumably invariable quantities, and two Schrödinger equations associated with equations (1) read as [6]

$$\begin{pmatrix}
\frac{\hbar^2}{2} & \Delta/2 + \varepsilon_c(p) \\
\Delta/2 - \varepsilon_c(p) & 0
\end{pmatrix}
\begin{pmatrix}
\Psi(x,t)
\end{pmatrix} = 0 .$$

The electron and hole states are correlated by charge and momentum balance and allow for a joint description in terms of the general dispersion law [5]

$$(E - E_c)(E - E_v) = \begin{pmatrix}
E - \frac{\Delta}{2} - \varepsilon_c(p)
\end{pmatrix}
\begin{pmatrix}
E + \left(\frac{\Delta}{2}\right) + \varepsilon_c(-p)
\end{pmatrix} = 0 .$$

For quadratic dispersion (3) reads as

$$(E - E_c)(E - E_v) = \begin{pmatrix}
E - \frac{\Delta}{2} - \frac{p^2}{2m_e}
\end{pmatrix}
\begin{pmatrix}
E + \left(\frac{\Delta}{2}\right) + \frac{p^2}{2m_h}
\end{pmatrix} = 0 .$$

The physical picture under (4) is that at very low electron density the Coulomb interaction between the electrons and ions and the phonons and ions is accounted for by effective mass of the conduction and valence states in the quadratic dispersion law $\varepsilon_{c,v} = p^2/(2m_{c,v})$. This effective mass approximation has substantial calculation advances as based on simple relations between the quadratic dispersion law and its quantized form.
In this section we follow [5] and emphasize specific features emerging beyond the effective mass and in the presence of the back reaction of phonons. The first quantized free-field equation of motion is given by relation

\[ \hat{E} - \frac{\Delta}{2} - \epsilon_i(\hat{\mathbf{p}}) \hat{E} + \left( \frac{\Delta}{2} \right) + \epsilon_i(\hat{\mathbf{p}}) \right] \Psi(x, t) = 0 \]  

where \( \Psi(x, t) \) is the two body wave function, \( \hat{E} = i \partial / \partial t \), and \( \hat{\mathbf{p}} = -i \nabla(\mathbf{x}) \).

The first quantized free-field equation of motion is given by relation

\[ \left\{ \left[ \hat{E} - \frac{\Delta}{2} - \epsilon_i(\hat{\mathbf{p}}) \right] \hat{E} + \left( \frac{\Delta}{2} \right) + \epsilon_i(\hat{\mathbf{p}}) \right\} \Psi(x, t) = 0 \],

where \( \Psi(x, t) \) is obeying the kinetic equation

\[ \left\{ \left[ -\frac{\partial^2}{\partial t^2} + i \frac{\partial}{\partial t} \right] \epsilon_i(-\hat{\mathbf{p}}) - \left( \frac{\Delta}{2} \right) - 2 \epsilon_i(-\hat{\mathbf{p}}) - \epsilon_i(\hat{\mathbf{p}}) \left( i \frac{\partial}{\partial t} \right) - \epsilon_i(\hat{\mathbf{p}}) \right\} \Psi(x, t) = 0 \].

Path to the relativistic Lagrange-Hamilton description starts with transformation to the oscillator-like equation having uniform differential form obtained by the phase transformation toward the auxiliary function \( \epsilon_i(t, x) \)

\[ \epsilon_i(t, x) = \exp \left\{ \frac{i}{2} \left[ A(\hat{\mathbf{p}}) \right] \right\} \Phi(x, t) \].

The oscillator-like equation reads as

\[ \partial_0 \Phi^*(x, t) \partial_0 \Phi(x, t) - \Omega^2(\hat{\mathbf{p}}) \Phi^*(x, t) \Phi(x, t) = 0 \]

and the effective frequency operator

\[ \Omega(\hat{\mathbf{p}}, t) = \frac{1}{2} \left[ \epsilon_i(-\hat{\mathbf{p}}) + \epsilon_i(\hat{\mathbf{p}}) + \Delta \right]. \]

With complex Klein-Gordon field in mind the Lagrangian density for the auxiliary function \( \Phi(x, t) \) is given by

\[ \mathcal{L}(x, t) = \frac{1}{\Delta} \left[ \partial_0 \Phi(x, t) \partial_0 \Phi^*(x, t) - \Omega^2(\hat{\mathbf{p}}) \Phi(x, t) \Phi^*(x, t) \right] \]

The factor \( 1 / \Delta \) proceed from the specific correspondence principle: at \( \Delta \rightarrow \infty \) the derived relations turn into the ordinary quantum mechanical analogies for independent bands. The total field function \( \Psi(x, t) \) is restored by inverse phase transformation

\[ \Phi(x, t) = \exp \left\{ - \frac{i \epsilon_i(\hat{\mathbf{p}}) - \epsilon_i(\hat{\mathbf{p}})}{2} \right\} \Psi(x, t) \]

As a result the Lagrangian density for the field function \( \Psi(x, t) \) reads as

\[ \mathcal{L}(x, t) = \frac{1}{\Delta^2} \left[ \left( i \epsilon_i(\hat{\mathbf{p}}) - \epsilon_i(\hat{\mathbf{p}}) \right) \Phi^*(x, t) + \Phi^*(x, t) \right] + \Omega^2(\hat{\mathbf{p}}) \Psi^2 \]

The derived entities are canonical moments.
\[
\pi = \frac{\partial L}{\partial \Psi} = -\frac{i(e_i(\hat{p}) - e_i(\hat{p})))}{2\hbar} \psi'(x, t) + \psi'(x, t), \quad (14)
\]

\[
\pi^* = \frac{\partial L}{\partial \Psi^*} = +\frac{i(e_i(\hat{p}) - e_i(\hat{p})))}{2\hbar} \psi(x, t) + \psi(x, t), \quad (15)
\]

and the free Hamiltonian density
\[
\mathcal{H} = \pi \psi^* + \pi^* \psi^* - L. \quad (16)
\]

Expansion of the Lagrangian density (13)
\[
L(\Psi, \Psi^*; \psi, \psi^*) = \frac{1}{\Delta} \left[ \frac{(e_i(\hat{p}) - e_i(\hat{p}))}{4} \psi \psi^* + \frac{i(e_i(\hat{p}) - e_i(\hat{p})))}{2} \psi' \psi^* - \frac{i(e_i(\hat{p}) - e_i(\hat{p})))}{2} \psi' \psi^* - \Omega^2(\hat{p}) \psi \psi^* \right] \quad (17)
\]

exhibits spatial derivative terms higher than second order and creates a self-interacting potential supplementary to the canonical Klein-Gordon form. The corresponding condition for the self-interacting potential being small is \( e_{\psi, \psi'} \ll \Delta \) with presumably holds for the energy of radiation of the order of band gap.

3. Decomposition of field functions for e-h pairs

Decomposition is the first step toward the derivation of field functions. Unlike the free Klein-Gordon particle constrained only by relativistic requirements, the particle in lattice environment is constrained by e-h dispersion relations. Into plane wave basis [5]
\[
\Psi(x, t) = \frac{1}{\sqrt{\nu}} \sum_p \int dE \tilde{\Psi}(E, p) e^{-iEt + ipx} \quad (18)
\]
the decomposition is defined on the hypersurfaces specified by the roots of (3), \( p \) is the crystal quasi-momentum, \( p_n = (2\pi/L)\nu_k \), \( n_k = 0,1,2,... \), and \( Et - p \cdot x = px \) is 4-vector scalar product. Within this vector field approach both direct and complex conjugate field functions applay as
\[
\Psi(x, t) = \frac{1}{\sqrt{\nu}} \sum_p \left( \int dE \tilde{\Psi}(E, p) e^{-iEt} \right) e^{ipx},\quad (19)
\]
\[
\Psi^*(x, t) = \frac{1}{\sqrt{\nu}} \sum_p \left( \int dE \tilde{\Psi}^*(E, p) e^{iEt} \right) e^{-ipx}. \quad (20)
\]

While the general dispersion law (3) is defined on grid, we not introduce new notations and write the Fourier transformation as
\[
\tilde{\Psi}(E, p) = \delta \left[ E - \frac{\Delta}{2} - \epsilon_i(p) \right] \left[ E + \frac{\Delta}{2} + \epsilon_i(-p) \right] \psi(E, p) \delta(g(E, p)) \psi(E, p), \quad (21)
\]
\[
\tilde{\Psi}^*(E, p) = \delta \left[ E - \frac{\Delta}{2} - \epsilon_i(p) \right] \left[ E + \frac{\Delta}{2} + \epsilon_i(-p) \right] \psi^*(E, p) \delta(g(E, p)) \psi^*(E, p). \quad (22)
\]

Inserting relations (21), (22) into the (19), (20) the field functions read as
\[
\Psi(x, t) = \frac{1}{\sqrt{\nu}} \sum_p \left( \int_{-\infty}^{\infty} dE \delta (g(E, p)) f(E, p) \right) e^{ipx}, \quad (23)
\]
\[
\Psi^*(x, t) = \frac{1}{\sqrt{\nu}} \sum_p \left( \int_{-\infty}^{\infty} dE \delta (g(E, p)) f^*(E, p) \right) e^{-ipx}. \quad (24)
\]
where \( f(E,p) = \psi(E,p)e^{-iEt} \) and \( f^*(E,p) = \psi^*(E,p)e^{iEt} \). The delta function in the integrands in (23), (24), formally is defined as a distribution for the function, and the integral in (19) coincides on the integral form of generalized scaling property. Harmonization of field functions with general dispersion (3) and the transition to field functions in operator representation is given in Appendix A (A27), (A28):

\[
\hat{\Psi}(x,t) = \frac{(2\pi)^{3/2}}{V} \sum_p \left[ \left( \frac{\Delta}{2\Omega(p)} + \hat{a}_p(t) + \hat{a}^*_p(-p,t) \right) \right] e^{ipx}, \tag{25}
\]

\[
\hat{\Psi}^*(x,t) = \frac{(2\pi)^{3/2}}{V} \sum_p \left[ \left( \frac{\Delta}{2\Omega(p)} - \hat{a}^*_p(t) + \hat{a}_p(-p,t) \right) \right] e^{-ipx}. \tag{26}
\]

In these terms the time derivatives of field functions reads in operator representation as:

\[
\hat{\dot{\Psi}}(x,t) = \frac{(2\pi)^{3/2}}{V} \sum_p \left[ \left( \frac{\Delta}{2\Omega(p)} - i(\Delta/2 + \varepsilon_e(p))\hat{a}_p(t) + i(\Delta/2 + \varepsilon_e(p))\hat{a}^*_p(-p,t) \right) \right] e^{ipx}, \tag{27}
\]

\[
\hat{\dot{\Psi}}^*(x,t) = \frac{(2\pi)^{3/2}}{V} \sum_p \left[ \left( \frac{\Delta}{2\Omega(p)} - i(\Delta/2 + \varepsilon_e(p))\hat{a}^*_p(t) - i(\Delta/2 + \varepsilon_e(p))\hat{a}_p(-p,t) \right) \right] e^{-ipx}. \tag{28}
\]

Calculations, associated with the principle of least action starts with the free Hamiltonian

\[
\mathcal{H}_{free} = \pi^2\dot{\Psi} + \pi^*\dot{\Psi}^* - \mathcal{L}. \tag{29}
\]

The total Hamiltonian is obtained from free Hamiltonian inserting dynamic dispersion in field function and canonical momentum terms by replacement \( p \rightarrow P = p - qA \). The 4-potential \( A = \{0,0,0,A_\tau\} \) corresponds the electromagnetic radiation approximated by electric field \( \varepsilon = -\dot{A} \). As a result the Lagrangian density reads as

\[
\mathcal{L} = \pi^2\dot{\Psi} + \pi^*\dot{\Psi}^* - \mathcal{H}_{tot}. \tag{30}
\]

In quasiparticle representation the Lagrangian density (30) comprises products of field functions which are complex conjugate in the space coordinate and that, in total, become functions on field operators. Their values are found by variational derivatives of the action \( \delta S = 0 \) [6]. The corresponding differential equations for field operators (A23)-(A26) are Heisenberg-like, first order in time derivative, and comprise time dependent coefficients so turning the problem to conventional algebra [8 (Eqs.(32-36))].

Going back to the electron-hole problem in presence of electric field it is convenient to go to the dynamic dispersion. Replacement the dynamic dispersion contributes to the dispersion laws \( \varepsilon_e(p) \rightarrow \varepsilon_e(P,(t)) \) and \( \varepsilon_v(p) \rightarrow \varepsilon_v(P,(t)) \), and it means that the hypersurfaces (1) becomes time dependent. The effect of dynamic dispersion on field operators (A23)-(A26) appears through transition amplitudes proportional to the ratio \( \dot{\Omega}/\Omega \) and turns to zero at \( A = 0 \).

4. Electron-phonon and hole-phonon interaction

The two band model, reconsidered in sections 2 and 3 is based on the concept of lattice potential as a source of the band structure and the e-h pairs emerging only under the electromagnetic radiation. Interaction of electrons and holes with the ionic lattice vary their kinetics and is categorized as the back – reaction of phonons. Phonons excited due the thermal bath at finite temperatures are omitted in this manuscript.

The electronic part of this back reaction is derived in [7,8] as sums over creation and annihilation operators derived by conventional quantum mechanics [9]. Propagation of the back reaction of phonons starts at the Lagrange – Hamilton stage as supplementary to the electromagnetic radiation. As a consequence, the back reaction contributes in the least action and, subsequently, in kinetic equations for the field operators.

In terms of conventional quantum mechanics (in this section the reduced Planck constant \( \hbar \) is restored) the system is distinguished by Hamiltonian:
\[ H = \sum_p E_e(p) a^+_e(p) a_e(p) + \sum_p E_h(p) a^+_h(p) a_h(p) + \sum_q \hbar \omega_q c^+_q c_q + \sum_{pq} V_{e(p+q)}(a^+_e(p) a^+_{e_p} a_{e_p} a^+_{e_p} \hat{q}_q + c^+_q a^+_{e_p} a_{e_p} a^+_{e_p} \hat{q}_q) \] (31)

where \( E_{e(h)}(p) \) is dispersion of electrons (holes) and \( V_{e(h)q} \) are matrix elements weighting the combinations of creation and annihilation operators. [9]. In the context of the back reaction, only the last two terms with mixed electron and hole operators are relevant. The mathematical technique transforming Eq.(14) into a pure electronic problem is Baker-Campbell-Hausdorff canonical transformation (BCH) [7] that gives following constituents of electron-phonon and hole-phonon interaction due to exchange of virtual phonons [8]:

\[ e = \sum_{p',q} V_{e(p+q)}^2 \left( \frac{1}{E_e(p') - E_e(p) - \hbar \omega_q} + \frac{1}{E_e(p) - E_e(p') + \hbar \omega_q} \right) a^+_{e_p} a^+_{e_p} a_{e_p} a_{e_p} c_q , \] (32)

\[ eh = \sum_{p',q} V_{e(p+q)} V_{h(p+q)} \left( \frac{1}{E_e(p') - E_e(p) - \hbar \omega_q} + \frac{1}{E_e(p) - E_e(p') + \hbar \omega_q} \right) a^+_{e_p} a^+_{e_p} a_{e_p} a_{e_p} c_{h_p} c_{h_p} , \] (33)

\[ h = \sum_{p',q} V_{e(p+q)}^2 \left( \frac{1}{E_h(p') - E_h(p) - \hbar \omega_q} + \frac{1}{E_h(p) - E_h(p') + \hbar \omega_q} \right) a^+_{h_p} a^+_{h_p} a_{h_p} a_{h_p} c_q , \] (34)

\[ he = \sum_{p',q} V_{h(p+q)} V_{e(p+q)} \left( \frac{1}{E_h(p') - E_h(p) - \hbar \omega_q} + \frac{1}{E_h(p) - E_h(p') + \hbar \omega_q} \right) a^+_{h_p} a^+_{h_p} a_{h_p} a_{h_p} c_{e_p} c_{e_p} . \] (35)

The underlying physics is that back reaction changes the kinetic energy of electron-hole system on equal grounds with the electromagnetic radiation. Formally it means that dynamic dispersion laws \( \varepsilon_e(p) \rightarrow \varepsilon_e(p(t)) \) and \( \varepsilon_h(p) \rightarrow \varepsilon_h(p(t)) \) turns to

\[ \varepsilon_e(p) \rightarrow \varepsilon_e(p(t)) + P_{e,int} \] (36)

and

\[ \varepsilon_h(p) \rightarrow \varepsilon_h(p(t)) + P_{h,int} \] (37)

where \( P_{e,int} = e + eh \), \( P_{h,int} = h + he \) are inputs calculated by nonrelativistic quantum mechanics [9].

5. Summary

Self-consistent description of electrons, holes and the associated phonons is addressed to the short time kinetics of interacting of electromagnetic radiation with matter and obeying requirements of quantum mechanics, quantum field theory and special relativity. Special attention is paid to the allowed time dependence of radiation which, unlike the existing models, is arbitrary. Another property crucial for high speed practical needs is the time response condensing full history of the material.

The state of art relies on well-known theoretical approaches to the free particles. With application to solids distinguished by the lattice of surrounding atoms these approaches become more complex and are not completely understood.

What is new in the presented research is extension of the model of noninteracting electrons and holes to the back reaction of phonons as based on disconnection of the electron – phonon and the electron hole – phonon interaction in purely electronic and hole parts, and putting extra terms in Lagrange – Hamilton approach. Formally these extra terms contribute on equal grounds with electromagnetic radiation with nonperturbative mathematical structure of the solution maintained.
The necessary inputs are available within nonrelativistic quantum mechanics and commercial codes while the key output is distribution function for electron – electron hole pair determining the observables of interest.

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**Appendix**
The delta function in the integrands in (23), (24), formally is defined as a distribution for the function, and coincides with the integral form of generalized scaling property written as

\[
\int_{-\infty}^{\infty} f(E, \mathbf{p}) \delta(g(E, \mathbf{p})) dE = \sum_{i} \frac{f(E_{i}, \mathbf{p})}{g'(E_{i})} = \frac{f(E_{i}, \mathbf{p})}{|g'(E_{i})|} + \frac{f(E_{2}, \mathbf{p})}{|g'(E_{2})|} \tag{A1}
\]

and

\[
\int_{-\infty}^{\infty} f^{*}(E, \mathbf{p}) \delta(g(E, \mathbf{p})) dE = \sum_{i} \frac{f^{*}(E_{i}, \mathbf{p})}{g'(E_{i})} = \frac{f^{*}(E_{1}, \mathbf{p})}{|g'(E_{1})|} + \frac{f^{*}(E_{2}, \mathbf{p})}{|g'(E_{2})|} \tag{A2}
\]

where

\[
g(E, \mathbf{p}) = \left[ E - \frac{\Delta}{2} - \varepsilon_{c}(\mathbf{p}) \right] \left[ E + \frac{\Delta}{2} + \varepsilon_{c}(\mathbf{p}) \right] \tag{A3}
\]

The roots of \( g(E, \mathbf{p}) \) read as

\[
E_{1} = \Delta/2 + \varepsilon_{c}(\mathbf{p}) \tag{A4}
\]
\[
E_{2} = (-\Delta/2 - \varepsilon_{c}(\mathbf{p})). \tag{A5}
\]

Derivative of \( g(E, \mathbf{p}) \) is equal to the derivative of dispersion relation (3)

\[
\frac{\partial g(E, \mathbf{p})}{\partial E} = 2E - \varepsilon_{c}(\mathbf{p}) + \varepsilon_{v}(\mathbf{p}) \tag{A6}
\]

Substituting (A4) and (A5) into (A6) gives

\[
g'(E_{1}) = (\Delta + \varepsilon_{c}(\mathbf{p}) + \varepsilon_{v}(\mathbf{p})) \tag{A7}
\]
\[
g'(E_{2}) = (-\Delta - \varepsilon_{c}(\mathbf{p}) - \varepsilon_{v}(\mathbf{p})) \tag{A8}
\]

and the common denominator

\[
|g'(E_{1})| = |g'(E_{2})| = (\Delta + \varepsilon_{c}(\mathbf{p}) + \varepsilon_{v}(\mathbf{p})) = 2\Omega(p) \tag{A9}
\]

Relation (A9) is the on shell condition for 4-field.

The numerators in (A1), (A2) start with \( f(E_{i}, \mathbf{p}) \quad i = 1, 2 \) function. For direct field functions

\[
f(E_{1}, \mathbf{p}) = \psi\left(\frac{\Delta}{2} + \varepsilon_{c}(\mathbf{p})\right) e^{-i\left(\frac{\Delta}{2} + \varepsilon_{c}(\mathbf{p})\right)^{\dagger}} = a_{c}(\mathbf{p}) e^{-i\left(\frac{\Delta}{2} + \varepsilon_{c}(\mathbf{p})\right)} \tag{A10}
\]
\[
f(E_{2}, \mathbf{p}) = \psi\left(-\frac{\Delta}{2} - \varepsilon_{c}(\mathbf{p})\right) e^{-i\left(-\frac{\Delta}{2} - \varepsilon_{c}(\mathbf{p})\right)^{\dagger}} = a_{h}(-\mathbf{p}) e^{-i\left(-\frac{\Delta}{2} - \varepsilon_{c}(\mathbf{p})\right)} \tag{A11}
\]

For complex conjugate field functions

\[
f^{*}(E_{1}, \mathbf{p}) = \psi^{*}\left(\frac{\Delta}{2} + \varepsilon_{c}(\mathbf{p})\right) e^{+i\left(\frac{\Delta}{2} + \varepsilon_{c}(\mathbf{p})\right)^{\dagger}} = a_{c}^{*}(-\mathbf{p}) e^{+i\left(\frac{\Delta}{2} + \varepsilon_{c}(\mathbf{p})\right)} \tag{A12}
\]
\[
f^* (E_2, \mathbf{p}) = \psi^* \left(-\frac{\Delta}{2} - \varepsilon_v (\mathbf{p})\right) e^{i\left(-\frac{\Delta}{2} - \varepsilon_v (\mathbf{p})\right)t} = a^*_h (\mathbf{p}) e^{i\left(-\frac{\Delta}{2} - \varepsilon_v (\mathbf{p})\right)t}.
\]  

(A13)

Going back to (A1) and (A2) and inserting (A9) – (A13) gives

\[
\Psi(x, t) = \frac{(2\pi)^{1/2}}{\sqrt{\nu}} \sum_p \left\{ \frac{1}{\sqrt{2 \Omega (\mathbf{p})}} \left( a_v (\mathbf{p}) e^{-i(\Delta + \varepsilon_v (\mathbf{p})t)} + a_h (\mathbf{p}) e^{i(\Delta + \varepsilon_v (\mathbf{p})t)} \right) \right\} e^{i\xi}.
\]  

(A14)

\[
\Psi^* (x, t) = \frac{(2\pi)^{1/2}}{\sqrt{\nu}} \sum_p \left\{ \frac{1}{\sqrt{2 \Omega (\mathbf{p})}} \left( a_v^* (\mathbf{p}) e^{i(\Delta + \varepsilon_v (\mathbf{p})t)} + a_h^* (\mathbf{p}) e^{-i(\Delta + \varepsilon_v (\mathbf{p})t)} \right) \right\} e^{-i\xi}.
\]  

(A15)

As it follows, the correspondence between effective frequency operator \( \Omega (\mathbf{p}) \) and the effective frequency \( \Omega (\mathbf{p}) = \frac{\Delta + \varepsilon_v (\mathbf{p}) + \varepsilon_v (\mathbf{p})}{2} \) valid for quadratic dispersion is maintained in case of arbitrary dispersion. As to the field amplitudes (A14), (A15), they are renormalized as

\[
\psi\left((\Delta/2 + \varepsilon_v (\mathbf{p})), \right) \rangle = a_v (\mathbf{p}),
\]  

(A16)

and similarly for other field operators. As a result, the field functions read as

\[
\Psi(x, t) = \frac{(2\pi)^{1/2}}{\sqrt{\nu}} \sum_p \left\{ \frac{1}{\sqrt{2 \Omega (\mathbf{p})}} \left( a_v (\mathbf{p}) e^{-i(\Delta + i\varepsilon_v (\mathbf{p})t)} + a_h (\mathbf{p}) e^{i(\Delta + i\varepsilon_v (\mathbf{p})t)} \right) \right\} e^{i\xi},
\]  

(A17)

\[
\Psi^* (x, t) = \frac{(2\pi)^{1/2}}{\sqrt{\nu}} \sum_p \left\{ \frac{1}{\sqrt{2 \Omega (\mathbf{p})}} \left( a_v^* (\mathbf{p}) e^{i(\Delta + i\varepsilon_v (\mathbf{p})t)} + a_h^* (\mathbf{p}) e^{-i(\Delta + i\varepsilon_v (\mathbf{p})t)} \right) \right\} e^{-i\xi}.
\]  

(A18)

Formally the field functions are redefined by dividing the factor \( \frac{1}{\sqrt{\Delta 2 \Omega (\mathbf{p})}} \) in (A17) and (A20) by the factor \( \frac{1}{\sqrt{\Delta 2 \Omega (\mathbf{p})}} \) resulting in

\[
\Psi(x, t) = \frac{(2\pi)^{1/2}}{\sqrt{\nu}} \sum_p \left\{ \frac{\Delta}{\sqrt{2 \Omega (\mathbf{p})}} \left( a_v (\mathbf{p}) e^{-i(\Delta + \varepsilon_v (\mathbf{p})t)} + a_h (\mathbf{p}) e^{i(\Delta + \varepsilon_v (\mathbf{p})t)} \right) \right\} e^{i\xi},
\]  

(A19)

\[
\Psi^* (x, t) = \frac{(2\pi)^{1/2}}{\sqrt{\nu}} \sum_p \left\{ \frac{\Delta}{\sqrt{2 \Omega (\mathbf{p})}} \left( a_v^* (\mathbf{p}) e^{i(\Delta + \varepsilon_v (\mathbf{p})t)} + a_h^* (\mathbf{p}) e^{-i(\Delta + \varepsilon_v (\mathbf{p})t)} \right) \right\} e^{-i\xi},
\]  

(A20)

Where \( \tilde{a}_v = (2 \Delta \Omega (\mathbf{p}))^{-1/2} a_v \), \( \tilde{a}_h = (2 \Delta \Omega (\mathbf{p}))^{-1/2} a_h \) and time derivatives from field functions (A19), (A20) read as (the punctuation ~ is omitted in further calculations):

\[
\dot{\Psi}(x, t) = \frac{(2\pi)^{1/2}}{\sqrt{\nu}} \sum_p \left\{ \frac{\Delta}{\sqrt{2 \Omega (\mathbf{p})}} \left( -i(\Delta + \varepsilon_v (\mathbf{p})t) a_v (\mathbf{p}) e^{-i(\Delta + \varepsilon_v (\mathbf{p})t)} + i(\Delta + \varepsilon_v (\mathbf{p})t) a_h (\mathbf{p}) e^{i(\Delta + \varepsilon_v (\mathbf{p})t)} \right) \right\} e^{i\xi},
\]  

(A21)

\[
\dot{\Psi}^* (x, t) = \frac{(2\pi)^{1/2}}{\sqrt{\nu}} \sum_p \left\{ \frac{\Delta}{\sqrt{2 \Omega (\mathbf{p})}} \left( -\frac{\Delta}{\sqrt{2 \Omega (\mathbf{p})}} a_v^* (\mathbf{p}) e^{i(\Delta + \varepsilon_v (\mathbf{p})t)} + \frac{\Delta}{\sqrt{2 \Omega (\mathbf{p})}} a_h^* (\mathbf{p}) e^{-i(\Delta + \varepsilon_v (\mathbf{p})t)} \right) \right\} e^{-i\xi}.
\]  

(A22)

Next concern is new time-dependent electron and hole operators:

\[
a_v (\mathbf{p}) e^{-i(\Delta + \varepsilon_v (\mathbf{p})t)} \Rightarrow \hat{a}_v (\mathbf{p}, t),
\]  

(A23)

\[
a_h (\mathbf{p}) e^{i(\Delta + \varepsilon_v (\mathbf{p})t)} \Rightarrow \hat{a}_h (\mathbf{p}, t),
\]  

(A24)

\[
a_v^* (\mathbf{p}) e^{i(\Delta + \varepsilon_v (\mathbf{p})t)} \Rightarrow \hat{a}_v^* (\mathbf{p}, t),
\]  

(A25)

\[
a_h^* (\mathbf{p}) e^{-i(\Delta + \varepsilon_v (\mathbf{p})t)} \Rightarrow \hat{a}_h^* (\mathbf{p}, t).
\]  

(A26)

Finally, the field functions in operator representation are derived from (A19), (A20) and reads as

\[
\hat{\Psi}(x, t) = \frac{(2\pi)^{1/2}}{\sqrt{\nu}} \sum_p \left\{ \frac{\Delta}{\sqrt{2 \Omega (\mathbf{p})}} (\hat{a}_v (\mathbf{p}, t) + \hat{a}_h (\mathbf{p}, t)) \right\} e^{i\xi},
\]  

(A27)

\[
\hat{\Psi}^* (x, t) = \frac{(2\pi)^{1/2}}{\sqrt{\nu}} \sum_p \left\{ \frac{\Delta}{\sqrt{2 \Omega (\mathbf{p})}} (\hat{a}_v^* (\mathbf{p}, t) + \hat{a}_h^* (\mathbf{p}, t)) \right\} e^{-i\xi}.
\]  

(A28)
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