Theory of the Two-Particle Emission from Superfluid Fermi Gases in the BCS-BEC Crossover

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Abstract

We present a theory of the emission of fermion pairs from a superfluid Fermi gas induced by a photon absorption. In the solid state physics, this type of process is called double photo-emission (DPE). The spectrum of the induced two-particle current (or DPE current) provides a direct insight into the pair-correlation of condensate fermion pairs. We develop a general formalism for two-particle current induced by DPE by treating the coupling of two Fermi gases with the time-dependent perturbation theory. This formalism is used to calculate energy distributions of DPE current from the superfluid Fermi gas in the BCS-BEC crossover at $T = 0$. We show that the DPE current has distinct contributions of the condensed pair components and uncorrelated pair states. We also calculate the angular dependence of DPE current in the BCS-BEC crossover. The DPE current of the tightly-bound molecules in the BEC regime is found to be quite different from that of the weakly-bound Cooper pairs.

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I. INTRODUCTION

In recent experiments of ultracold atomic Fermi gases, the crossover from the BCS-type superfluid to the Bose-Einstein condensation (BEC) of tightly bound molecules including the unitary gas as an intermediate regime have been realized using a tunable pairing interaction associated with a Feshbach resonance [1–3]. While a lot of studies are concerned with the thermodynamic properties of superfluidity as well as collective modes in ultracold atomic gases [4], the basic properties of condensate atom pairs is also interesting problem from the conceptual viewpoint. In particular, quasiparticle excitations were studied by using the momentum-resolved photoemission-type spectroscopy, in which atoms are transferred to the third empty atomic state by rf pulse [5]. This powerful technique, which is an analog of the angle-resolved photoemission spectroscopy (ARPES) in solid state physics, allows one to measure microscopic properties of a cold Fermi gas in the crossover region. The photoemission-type spectroscopy has been theoretically addressed in the literature of cold Fermi gas [6, 7]. The experimental results are in qualitative agreement with the theoretical calculation.

In this paper, we consider an alternative approach to study the basic properties of condensate atom pairs in a superfluid Fermi gas. The main purpose of this paper is to distinguish between cooper pairs and tightly bound molecules in the BCS-BEC crossover from the emission of Fermi condensate pair. In solid state physics, emission of election pair induced by photon absorption is called the double photoemission (DPE) and is used to measured two-particle spectra that provide direct insight into the energy and the angular dependence of the pair-correlation functions [8, 9]. Many experiments have observed DPE spectroscopy from conventional and unconventional superconducting samples [10–12]. In view of the advances in experimental techniques in ultracold atomic gases, an analogous experiment on Fermi atomic superfluid may be expected to become available. In this paper, we provides a general theory of two-particle current induced by DPE (DPE current) from superfluid Fermi gases [13, 14]. We note that the previous studies of the photoemission-type spectroscopy [6, 7] essentially deals with the situation where a single atom is emitted from the system by absorbing a photon. In the present paper, we consider a possibility of emission of a pair of atoms by photon absorption. As an illustration, we consider the BCS-BEC crossover at $T = 0$ within the framework of BCS-Leggett’s theory. We calculate the energy dependence
of DPE current, and explicitly show that the contribution of uncorrelated pair states and that of condensed pairs are clearly separated. We will also show that the angular dependence of DPE current, which indicate the contribution of condensed pairs of DPE current see clear compared to single-photoemission current.

In Sec. II, we will derive a general expression for DPE current of atoms tunneling between two atomic gases coupled through an external field. By treating the coupling between two fermi gases as perturbation, we employ time-dependent perturbation theory up to forth order in order to obtain non-vanishing contribution to two-particle current. We will then introduce a two-particle spectral function describing DPE current.

In Sec. III, we will calculate the two-particle spectral function in the BCS-BEC crossover at $T = 0$ using Leggett’s theory [14] based on the mean-field treatment.

In Sec. IV, we will show the calculations of DPE current as a function of the energy transfer from coupling field, and discuss the separate contributions of the condensed pair components and uncorrelated pair states. We will also show the angular distributions of DPE current and discuss the possibility to distinguish between Cooper pairs and molecules form the calculations of DPE current.

For comparison, we will also show the single-particle current in the BCS-BEC crossover in Sec. V including the higher-order process involving two-particle tunneling. We will show the contribution of uncorrelated pair states is much lager than that of condensed pairs, although there appears a small peak from contribution of condensed pairs.

II. GENERAL FORMALISM FOR TWO-PARTICLE CURRENT OF A TWO-COMPONENT FERMI GAS

In this section, we present a formalism for two-particle current induced by DPE by treating the coupling of two Fermi gases with the time-dependent perturbation theory. In Fig. II we illustrate the emission of fermion pairs induced by a photon absorption from a superfluid Fermi gas. We assume that atoms are initially in state 1 and transferred to the state 2 when a coupling interaction is switched on. The tunneling perturbation $V(t)$ couples the two many-body systems together by introducing a mechanism by which an atom can tunnel between the two systems. This perturbation has terms that create an atom in one system while destroying an atom in the other system and vice versa.
The total Hamiltonian for a two-component Fermi gas is given by

$$\hat{H} = \hat{H}_1 + \hat{H}_2 + \hat{V}(t) \equiv \hat{H}_0 + \hat{V}(t),$$

where $\hat{H}_1$ describes the initial state of interest, and $\hat{H}_2$ describes the final state that is coupled to the state 1 through the tunneling Hamiltonian $\hat{V}(t)$ given by

$$\hat{V}(t) = \sum_{\sigma} \sum_k e^{\eta t} (\gamma_{\sigma} e^{-i\omega_{\sigma} t} \hat{b}_{k+q_{\sigma}}^\dagger \hat{c}_{k\sigma} + \text{H.c.}).$$

Here $\hat{c}_{k\sigma}$ and $\hat{b}_{k\sigma}$ are creation operators of the state 1 and 2. $\omega_{\sigma}$ and $q_{\sigma}$ are the effective energy transfer and the momentum transfer from the coupling fields, and $\gamma_{\sigma}$ is the coupling strength. We have also introduced a factor $e^{\eta t}$ ($\eta > 0$) that models the adiabatic switching on of the interaction at $t \to -\infty$.

**A. Two-Particle Current**

Let us define the two-particle density for the state 2 as

$$n(k_1 \uparrow, k_2 \downarrow, t) \equiv \langle \hat{b}_{k_1 \uparrow}^\dagger \hat{b}_{k_2 \downarrow}^\dagger \hat{b}_{k_2 \downarrow} \hat{b}_{k_1 \uparrow} \rangle_t \equiv \langle \hat{n}(k_1 \uparrow, k_2 \downarrow) \rangle_t,$$

where

$$\hat{n}(k_1 \uparrow, k_2 \downarrow) \equiv \hat{b}_{k_1 \uparrow}^\dagger \hat{b}_{k_2 \downarrow}^\dagger \hat{b}_{k_2 \downarrow} \hat{b}_{k_1 \uparrow}.$$  

This two-particles density describes the density of fermion pairs whose momentum $k_1$ and $k_2$.

Two-particle current is defined by time derivative of the two-particle density

$$J(k_1 \uparrow, k_2 \downarrow) = \frac{d}{dt} n(k_1 \uparrow, k_2 \downarrow) = \frac{i}{\hbar} \langle [\hat{n}(k_1 \uparrow, k_2 \downarrow, t), \hat{H}] \rangle_t.$$

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FIG. 1. (Color online) The illustration for emission of fermion pairs from two-particle induced by a photon absorption from a superfluid Fermi gas.
This two-particle current describe the number of atom pairs emitted per unit time. One can easily show that \( \hat{H}_1 \) and \( \hat{H}_2 \) make no contribution to the commutator in the r.h.s. of Eq. (5) and thus

\[
J(\mathbf{k}_1 \uparrow, \mathbf{k}_2 \downarrow) = \frac{i}{\hbar} \left\langle \left[ \hat{n}(\mathbf{k}_1 \uparrow, \mathbf{k}_2 \downarrow), \hat{V}(t) \right] \right\rangle_t .
\]  

(6)

Using the expression (2) for the tunneling Hamiltonian, we find that the two-particle current is given by

\[
J(t) = e^{\eta t} \left[ \gamma_\uparrow e^{-i\omega_\uparrow t} \left\langle \hat{F}_\uparrow(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_1 - \mathbf{q}_\uparrow) \right\rangle_t + \gamma_\downarrow e^{-i\omega_\downarrow t} \left\langle \hat{F}_\downarrow(\mathbf{k}_2, \mathbf{k}_1, \mathbf{k}_2 - \mathbf{q}_\downarrow) \right\rangle_t - \text{c.c.} \right] ,
\]

(7)

where we have introduced the following four field correlation function:

\[
\hat{F}_\uparrow(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4) \equiv \hat{b}_{k_1 \uparrow}^{\dagger} \hat{b}_{k_2 \downarrow}^{\dagger} \hat{b}_{k_3 \downarrow} \hat{c}_{k_4 \uparrow}, \quad \hat{F}_\downarrow(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4) \equiv \hat{b}_{k_1 \downarrow}^{\dagger} \hat{b}_{k_2 \uparrow}^{\dagger} \hat{b}_{k_3 \uparrow} \hat{c}_{k_4 \downarrow} .
\]

(8)

Therefore, the two-particle current is expressed in terms of the correlation functions \( \langle \hat{F}_\uparrow \rangle \) and \( \langle \hat{F}_\downarrow \rangle \). In the next section, we employ the time-dependent perturbation theory to derive expressions for these correlation functions.

**B. General formalism of time evolution**

In general, nonequilibrium statical average of an arbitrary operator \( \hat{O} \) is given by

\[
\langle \hat{O} \rangle = \text{Tr} \{ \hat{\rho}(t) \hat{O} \} = \text{Tr} \{ \hat{U}(t, t_0) \hat{\rho}_0(t_0) \hat{U}^\dagger(t, t_0) \hat{O} \} = \text{Tr} \{ \hat{\rho}(t_0) \hat{O}_H(t) \} ,
\]

(9)

where \( \hat{\rho}(t) \) is the nonequilibrium statistical density operator, \( t_0 \) is the initial time, and \( \hat{U}(t, t_0) \) is the time evolution operator,

\[
\hat{U}(t, t_0) = \mathcal{T} \exp \left[ -\frac{i}{\hbar} \int_{t_0}^{t} dt' \hat{H}(t') \right] ,
\]

(10)

\( \mathcal{T} \) being time-ordering operator. \( \hat{O}_H \) is the Heisenberg operator denied by

\[
\hat{O}_H(t) = \hat{U}(t, t_0) \hat{O} \hat{U}(t, t_0) .
\]

(11)

In order to perform perturbative expansion in the tunneling Hamiltonian \( \hat{V}(t) \), we introduce the Heisenberg operator with respect to \( \hat{H}_0 \) as

\[
\hat{O}_{\hat{H}_0}(t) \equiv e^{i\hat{H}_0(t-t_0)/\hbar} \hat{O} e^{-i\hat{H}_0(t-t_0)/\hbar} .
\]

(12)
Then, $\hat{O}_H$ and $\hat{O}_{H_0}$ are related though the unitary transformation
\begin{equation}
\hat{O}_H(t) = \hat{U}(t, t_0) \hat{O}_{H_0}(t) \hat{U}(t, t_0).
\end{equation}

where
\begin{equation}
\hat{U}(t, t_0) = \mathcal{T} \exp \left[ -\frac{i}{\hbar} \int_{t_0}^{t} dt' \hat{V}_{H_0}(t') \right],
\end{equation}

Therefore, we find
\begin{equation}
\langle \hat{O} \rangle_t = \langle T \exp \left[ -\frac{i}{\hbar} \int_{t_0}^{t} dt' \hat{V}_{H_0}(t') \right] \hat{O}_{H_0}(t_0) \mathcal{T} \exp \left[ \frac{i}{\hbar} \int_{t_0}^{t} dt' \hat{V}_{H_0}(t') \right] \rangle_{t_0}.
\end{equation}

where $\mathcal{T}$ is the anti-chronological time-ordering operator.

The above expressions are conveniently denoted by using the Keldysh contour-time-path description \[15\]. In general, the nonequilibrium expectation value of the physical quantity can be written as (we omit the subscript $H$ for simplicity)
\begin{equation}
\langle \hat{O} \rangle_t = \langle \mathcal{T_c} \left[ \exp \left( -\frac{i}{\hbar} \oint_{t_0}^{t} dt' \hat{V}(t') \right) \right] \hat{O}(t) \rangle_{t_0},
\end{equation}

where $\mathcal{T}_c$ is a contour-ordering operator and
\begin{equation}
\oint_{t_0}^{t} = \int_{t_0}^{t} + \int_{t}^{t_0} = \int_{t_0}^{t} - \int_{t_0}^{t}
\end{equation}

stands for the integral along the contour. Expanding \[16\] in the perturbation Hamiltonian $\hat{V}$, we obtain the following expansion
\begin{equation}
\langle \hat{O} \rangle_t = \sum_{n=0}^{\infty} \langle \hat{O}^{(n)} \rangle_t,
\end{equation}

where the $n$-th order contribution is expressed as
\begin{equation}
\langle \hat{O}^{(n)} \rangle_t \equiv \frac{1}{n!} \left( \frac{i}{\hbar} \right)^n \oint_{t_0}^{t} dt_1 \cdots \oint_{t_0}^{t} dt_n \langle \mathcal{T} \left[ \hat{V}(t_1) \cdots \hat{V}(t_n) \hat{O}(t) \right] \rangle_{t_0}.
\end{equation}

C. General Expression for Two-Particle Current

We now consider the quantity $\langle F_a \rangle$ defined in Sec. II A. Under the assumption that the system 1 and 2 are uncoupled in the absence of the coupling Hamiltonian $V$, the first and second order contributions in the perturbative expansion \[19\] vanish. We thus left with the
third-order term as the lowest-order non-vanishing contribution. The expression for \( \langle \hat{F}_\sigma \rangle \) is then given by

\[
\langle \hat{F}_\tau (k_1, k_2, k_2, k_1 - q_1) \rangle^{(3)}_t = \left( -\frac{i}{\hbar} \right)^3 \gamma^\dagger \gamma |\gamma_0|^2 e^{i\omega_1 t} \int_{t_0}^{t_3} dt_1 \int_{t_0}^{t_2} dt_2 \int_{t_0}^{t_3} dt_3 e^{i(\omega_1 - \epsilon_{k_1}) / \hbar} e^{i(\omega_2 - \epsilon_{k_2}) / \hbar} e^{i(\omega_3 - \epsilon_{k_3}) / \hbar} \times \langle \hat{T} \left[ \hat{c}_{k_2-q_1}^{\dagger} (t_2) \hat{c}_{k_1-q_1}^{\dagger} (t_1) \right] \hat{c}_{k_1-q_1} (0) \hat{c}_{k_2-q_1} (t_3) \rangle_{t_0}, \quad (20)
\]

\[
\langle \hat{F}_\sigma (k_2, k_1, k_2 - q_1) \rangle^{(3)}_t = \left( -\frac{i}{\hbar} \right)^3 \gamma^\dagger \gamma |\gamma_0|^2 e^{i\omega_1 t} \int_{t_0}^{t_3} dt_1 \int_{t_0}^{t_2} dt_2 \int_{t_0}^{t_3} dt_3 e^{i(\omega_1 - \epsilon_{k_1}) / \hbar} e^{i(\omega_2 - \epsilon_{k_2}) / \hbar} e^{i(\omega_3 - \epsilon_{k_3}) / \hbar} \times \langle \hat{T} \left[ \hat{c}_{k_1-q_1}^{\dagger} (t_2) \hat{c}_{k_1-k_2}^{\dagger} (t_1) \right] \hat{c}_{k_1-q_1} (0) \hat{c}_{k_1-q_1} (t_3) \rangle_{t_0}, \quad (21)
\]

As we noted before, we assume that \( \eta |t| \ll 1 \).

Let us define the two-particle correlation functions by

\[
i G_\tau (k'_1, k'_2, t_1, t_2, t_3) \equiv \Theta (-t_1) \Theta (-t_2) \Theta (-t_3) \langle \hat{T} \left[ \hat{c}_{k'_2}^{\dagger} (t_2) \hat{c}_{k'_1}^{\dagger} (t_1) \right] \hat{c}_{k'_1} (0) \hat{c}_{k'_2} (t_3) \rangle_{t_0}, \quad (22)
\]

\[
i G_\sigma (k'_1, k'_2, t_1, t_2, t_3) \equiv \Theta (-t_1) \Theta (-t_2) \Theta (-t_3) \langle \hat{T} \left[ \hat{c}_{k'_1}^{\dagger} (t_1) \hat{c}_{k'_2}^{\dagger} (t_2) \right] \hat{c}_{k'_2} (0) \hat{c}_{k'_1} (t_3) \rangle_{t_0}. \quad (23)
\]

The Fourier transforms of these correlation functions are defined by

\[
G_\sigma (k_1, k_2, \omega_1, \omega_2, \omega_3) = \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \int_{-\infty}^{\infty} dt_3 e^{i\omega_1 t_1} e^{i\omega_2 t_2} e^{-i\omega_3 t_3} G_\sigma (k_1, k_2, t_1, t_2, t_3). \quad (24)
\]

Thus, the two-particle current is expressed in terms of these correlation functions as

\[
J (k_1, k_2) = -\frac{1}{\hbar} |\gamma|^2 |\gamma_0|^2 \left[ i G_\tau (k_1 - q_1, k_2 - q_4, \omega_1 - \epsilon_{k_1}, \omega_2 - \epsilon_{k_2}, \omega_3 - \epsilon_{k_2} + i\eta) + i G_\sigma (k_1 - q_1, k_2 - q_4, \omega_1 - \epsilon_{k_1}, \omega_2 - \epsilon_{k_2}, \omega_3 - \epsilon_{k_2} + i\eta) - c.c. \right]
\]

\[
= -\frac{2}{\hbar} |\gamma|^2 |\gamma_0|^2 \text{Im} \left[ G_\tau (k_1 - q_1, k_2 - q_4, \omega_1 - \epsilon_{k_1}, \omega_2 - \epsilon_{k_2}) + G_\sigma (k_1 - q_1, k_2 - q_4, \omega_1 - \epsilon_{k_1}, \omega_2 - \epsilon_{k_2}) \right], \quad (25)
\]

where we have denoted

\[
G_\tau (k_1 - q_1, k_2 - q_4, \omega_1 - \epsilon_{k_1}, \omega_2 - \epsilon_{k_2}) \equiv G_\tau (k_1 - q_1, k_2 - q_4, \omega_1 - \epsilon_{k_1} - i\eta, \omega_2 - \epsilon_{k_2} + i\eta), \quad (26)
\]

\[
G_\sigma (k_1 - q_1, k_2 - q_4, \omega_1 - \epsilon_{k_1}, \omega_2 - \epsilon_{k_2}) \equiv G_\sigma (k_1 - q_1, k_2 - q_4, \omega_1 - \epsilon_{k_1} + i\eta, \omega_2 - \epsilon_{k_2} - i\eta). \quad (27)
\]
D. Lehmann representation

In order to gain physical insight into the above results, it is useful to write down the expression (24) for the two-particle current in the Lehmann representation. The statistical average \( \langle \hat{O} \rangle_{t_0} \) of any operator can be expressed in terms of the energy eigenstates of \( \hat{H}_1 \) as

\[
\langle \hat{O} \rangle_{t_0} = \sum_n \rho_n \langle n | \hat{O} | n \rangle, \tag{28}
\]

where \( \rho_n \) is the diagonal element of the equilibrium statistical density operator in the energy representation. In the grand-canonical ensemble, it is given as

\[
\rho_n = \frac{\exp[-\beta(E_n - \mu N_n)]}{\Xi}, \quad \Xi \equiv \sum_n \exp[-\beta(E_n - \mu N_n)]. \tag{29}
\]

In the energy representation, the matrix element of the Heisenberg operator is given by

\[
\langle n | \hat{O}(t) | m \rangle = \langle n | e^{i \hat{H} t/\hbar} \hat{O} e^{-i \hat{H} t/\hbar} | m \rangle = e^{i(E_n - E_m)t/\hbar} \langle n | \hat{O} | m \rangle. \tag{30}
\]

Using this energy representation, we can express the two-particle correlation functions as

\[
G_{\uparrow}(k_1, k_2, \omega_1, \omega_2) + G_{\downarrow}(k_1, k_2, \omega_1, \omega_2) = -\sum_n \sum_l \frac{\rho_n}{\omega_1 + \omega_2 - (E_l - E_n)/\hbar - 2i\eta} \times \sum_m \frac{\langle l | \hat{c}_{k_1 \uparrow} | m \rangle \langle m | \hat{c}_{k_2 \downarrow} | n \rangle}{\omega_1 - (E_m - E_n)/\hbar + i\eta} - \sum_m \frac{\langle l | \hat{c}_{k_1 \uparrow} | m \rangle \langle m | \hat{c}_{k_2 \downarrow} | n \rangle}{\omega_2 - (E_m - E_n)/\hbar + i\eta}, \tag{31}
\]

We thus obtain the general expression for the two-particle current as

\[
J(k_1, k_2) = \frac{2\pi}{\hbar} \frac{1}{|\gamma_{\uparrow}|^2 |\gamma_{\downarrow}|^2} \sum_n \sum_l \rho_n \delta(\omega_1 + \omega_2 - (E_l - E_n)/\hbar) \times \left| \sum_m \frac{\langle l | \hat{c}_{k_1 \uparrow} | m \rangle \langle m | \hat{c}_{k_2 \downarrow} | n \rangle}{\omega_1 - (E_m - E_n)/\hbar + i\eta} - \sum_{m'} \frac{\langle l | \hat{c}_{k_1 \uparrow} | m' \rangle \langle m' | \hat{c}_{k_2 \downarrow} | n \rangle}{\omega_2 - (E_{m'} - E_n)/\hbar + i\eta} \right|^2, \tag{32}
\]

where

\[
\omega_1 = \omega_{\uparrow} - \epsilon_{k_1 \uparrow}, \omega_2 = \omega_{\downarrow} - \epsilon_{k_2 \downarrow}, k_1' = k_1 - q_{\uparrow}, k_2' = k_2 - q_{\downarrow}. \tag{33}
\]

The energy delta function in (32) arises from taking the limit \( \eta \to 0 \) in the common prefactor of (31).

The physical meaning of the energy eigenstates \( |n\rangle, |m\rangle, |m'\rangle, |l\rangle \) are understood as follows: \( |n\rangle \) describes the initial state, \( |m\rangle \) describes the state where a \( k_1' \uparrow \) particle is subtracted from the initial state, \( |m'\rangle \) describes the state where a \( k_2' \downarrow \) particle is subtracted from the initial state, and \( |l\rangle \) describes the state where \( k_1' \uparrow \) particle and \( k_2' \downarrow \) particle are subtracted from the initial state.
III. TWO-PARTICLE SPECTRUM IN FERMI SUPERFLUID

A. BCS-Leggett’s theory

We now consider a uniform Fermi superfluid gas at \( T = 0 \) to illustrate the most basic physics. In order to calculate the two-particle spectral function explicitly, we must specify a microscopic approximation. Here we use Leggett’s theory \([14]\) based on the mean-field treatment in the BCS-BEC crossover at \( T = 0 \). For this purpose, we work with the grand-canonical Hamiltonian defined by

\[
\hat{K}_1 = \hat{H}_1 - \mu \hat{N}_1, \tag{34}
\]

where \( \hat{N}_1 \equiv \sum_\sigma \sum_k \hat{c}_{k\sigma}^\dagger \hat{c}_{k\sigma} \) and \( \mu \) is the chemical potential of the system 1. It is convenient to introduce the Heisenberg operator defined in terms of the grand canonical Hamiltonian as

\[
\hat{c}_{k\sigma K}(t) \equiv e^{i\hat{K}_1 t/\hbar} \hat{c}_{k\sigma} e^{-i\hat{K}_1 t/\hbar} = e^{i\hat{H}_1 t/\hbar} e^{-i\mu \hat{N}_1 t/\hbar} \hat{c}_{k\sigma} e^{i\mu \hat{N}_1 t/\hbar} e^{-i\hat{H}_1 t/\hbar}. \tag{35}
\]

Here we have used the fact that the Hamiltonian commutes with the total number operator. Using the identity \( e^{-i\mu \hat{N}_1 t/\hbar} \hat{c}_{k\sigma} e^{i\mu \hat{N}_1 t/\hbar} = e^{i\mu t/\hbar} \hat{c}_{k\sigma} \), it is easily verified that

\[
\hat{c}_{k\sigma H}(t) = e^{i\mu t/\hbar} \hat{c}_{k\sigma H}(t) \quad \text{or} \quad \hat{c}_{k\sigma H}(t) = e^{-i\mu t/\hbar} \hat{c}_{k\sigma K}(t). \tag{36}
\]

The grand-canonical Hamiltonian for a uniform Fermi superfluid is given by

\[
\hat{K}_1 = \sum_{k,\sigma} \xi_k \hat{c}_{k\sigma}^\dagger \hat{c}_{k\sigma} + U \sum_{k,k',q} \hat{c}_{k+q/2\uparrow}^\dagger \hat{c}_{-k'+q/2\downarrow}^\dagger \hat{c}_{-k'+q/2\downarrow} \hat{c}_{k+q/2\uparrow}, \tag{37}
\]

where \( \xi_k = \epsilon_k - \mu \) with \( \epsilon_k = \frac{\hbar k^2}{2m} - \mu \). The pairing interaction \( U = -\frac{4\pi \hbar^2 a_s}{m} \) is assumed to be tunable by a Feshbach resonance, which is related to the s-wave scattering length \( a_s \). We introduce the mean-field static superfluid order parameter \( \Delta \) as,

\[
\Delta = U \sum_k \langle \hat{c}_{k\downarrow} \hat{c}_{k\uparrow} \rangle. \tag{38}
\]

As usual, we define the quasiparticle operators by the Bogoliubov transformation:

\[
\hat{c}_{k\uparrow} = u_k \hat{\alpha}_{k\uparrow} + v_k \hat{\alpha}_{-k\downarrow}; \quad \hat{c}_{k\downarrow} = u_k \hat{\alpha}_{k\downarrow} - v_k \hat{\alpha}_{-k\uparrow}. \tag{39}
\]
In terms of these quasiparticle operators, the grand canonical Hamiltonian can be written as

\[ \hat{K}_1 = \sum_k E_k (\hat{\alpha}_{k\uparrow} \hat{\alpha}_{k\uparrow} + \hat{\alpha}_{k\downarrow} \hat{\alpha}_{k\downarrow}) + \text{constant}, \]  

(40)

where the quasiparticle energy is given by

\[ E_k = \sqrt{\left( \frac{\hbar^2 k^2}{2m} - \mu \right)^2 + \Delta^2}. \]  

(41)

Denoting the Heisenberg operator defined by \( \hat{K}_1 \) as \( \hat{\alpha}_{k\sigma}(t) \), we have

\[ \hat{\alpha}_{k\sigma}(t) = \hat{\alpha}_{k\sigma} e^{-iE_k t/\hbar}, \quad \hat{\alpha}_{k\sigma}^\dagger(t) = \hat{\alpha}_{k\sigma} e^{iE_k t/\hbar}. \]  

(42)

In the usual (weak-coupling) BCS theory, the chemical potential \( \mu \) can be taken to be equal to the Fermi energy \( \epsilon_F \). However, from the general point of view, \( \mu \) should be determined by the equation for the number of fermions. Indeed, the chemical potential is found to remarkably deviate from the Fermi energy when the pairing interaction is strong. Within Leggett’s theory, the gap function \( \Delta \) and chemical potential \( \mu \) are determined self-consistently from the following gap and number equations for a uniform Fermi gas.

\[ 1 = -\frac{4\pi a_s}{m} \sum_k \left[ \frac{1}{2E_k} - \frac{1}{\epsilon_k} \right], \]  

(43)

\[ N = \sum_k \left[ 1 - \frac{\xi_k}{E_k} \right]. \]  

(44)

Assuming the BCS ground state that satisfies \( \hat{\alpha}_{k\sigma}|\Phi_0\rangle = 0 \), it is now straightforward to calculate the two particle correlation functions (22) and (23). The final result for the two-particle current is given by

\[ J(k_1, k_2, \omega'_1, \omega'_2) = \frac{2}{\hbar^2} |\gamma| |\gamma'|^2 \text{Im} \left[ \frac{v_{k_1}^2 u_{k_1'}^2}{\omega'_1 + \omega'_2 + 2\mu/\hbar - 2i\eta} \delta_{k'_1 - k'_2} \right. \]

\[ + \frac{v_{k_2}^2 u_{k_2'}^2}{\omega'_1 + \omega'_2 - (E_{k'_1} + E_{k'_2} - 2\mu)/\hbar - 2i\eta} \left. \right] \times \frac{1}{\omega'_1 - (E_{k'_1} - \mu)/\hbar - i\eta} + \frac{1}{\omega'_2 - (E_{k'_2} - \mu)/\hbar - i\eta}, \]  

(45)

where

\[ k'_1 = k_1 - q_{\uparrow}, \quad k'_2 = k_2 - q_{\downarrow}, \quad \omega'_1 = \omega_{\uparrow} - \epsilon_{k_1}/\hbar, \quad \omega'_2 = \omega_{\downarrow} - \epsilon_{k_2}/\hbar. \]  

(46)

In Eq. (45), the first term represents the contribution from the condensed pair components, while the second term is contribution from the uncorrelated pair states.
B. The two-particle spectral function in the BCS-BEC crossover

Hereafter we assume the case of $\omega^\uparrow = \omega^\downarrow = \omega$ for simplicity. Figure 2 shows intensity of the two-particle current $J(k_1, k_2, \omega'_1, \omega'_2)$ for $k'_1 = -k'_2$. The peak at $\hbar \omega = 0$ is the contribution of the condensed pair components, while the peak at $\hbar \omega \neq 0$ is the contribution of uncorrelated pair states. The weight of condensed pair contribution increases with increasing pairing interaction.

Figure 2. (Color online) Intensity of two-particle current. The values of the pairing interaction $(k_F a_s)^{-1}$ are (a) -1, (b) 0, and (c) 1.

Figure 3 shows the energy distribution of DPE current defined by $J(\omega) \equiv \sum_{k_1, k_2} J(k_1, k_2, \omega)$. 
FIG. 3. DPE current as a function of the energy $\omega$. The values of the pairing interaction $(k_F a_s)^{-1}$ are (a) -1, (b) 0, and (c) 1.

FIG. 4. Sketch of DPE process indicating two outgoing Fermions of with wave vector $k_1$ and $k_2$ as well as the emission angles $\theta_1$ and $\theta_2$.

We can see that the weight of condensed pair contribution increases with increasing pairing interaction. As we will see in Sec. IV, such a condensed pair contribution does not appear in the single-particle spectrum even when considering the two-particle tunneling contribution. In contrast, DPE current as a function of the energy exhibits distinct contributions of uncorrelated pair states and condensed pair components. However, in order to distinguish between weakly-bound Cooper pairs and tightly-bound molecules, it is not sufficient to see
only the energy distributions of DPE current. From this point of view, we will show the angular distributions of DPE current.

We define the angular distribution of DPE current as $J(\theta, \omega) = \sum_{\mathbf{k}_1, \mathbf{k}_2} J(\mathbf{k}_1, \mathbf{k}_2, \omega) \delta(\theta_2 - \theta_1 - \theta)$, where $\theta_1$ and $\theta_2$ are the scattering angles of the emitted pair of fermions and $\theta$ is the relative scattering angle, as illustrated in Fig. 4.

In Fig. 5, we plot the angular distribution of DPE current for $\omega = 0$. In the BCS side ($1/k_F a_s = -1$), we see the double peak of DPE current, which corresponds to the case where the particles constructing a pair are respectively emitted. On the contrary, in the BEC side ($1/k_F a_s = 1$) the single peak appears, which means that the pairs emitted as molecules. This shows that one can distinguish between Cooper pairs and molecules from the angular distributions of DPE current. We will show in the next section that such a condensed pair contribution cannot be seen by single-particle spectroscopy even when including the effect of the two-particle tunneling as the higher-order process in the tunneling Hamiltonian.

**IV. EFFECT OF TWO PARTICLE TUNNELING IN THE SINGLE-PARTICLE CURRENT**

In the case of superconductivity, effect of two-electron tunneling to single-particle current (i.e. Josephson current) was discussed by using the forth order perturbation theory. In this section, we give an analogous discussion of how the two-particle tunneling process affect the single-particle current in a superfluid Fermi gas in BCS-BEC crossover. In order to include the effect of two-particle tunneling, we have to calculate the forth order contribution.
A. General Formalism

The single-particle current is defined as the rate of change of the single-particle density

\[ n_\sigma(k, t) = \langle \hat{b}_k^{\dagger} \hat{b}_{k\sigma} \rangle_t, \tag{47} \]

which is given as

\[ J_\sigma = \frac{d}{dt} n_\sigma(k, t) = \frac{i}{\hbar} \langle \{ \hat{b}_k^{\dagger} \hat{b}_{k\sigma}, \hat{H} \} \rangle_t. \tag{48} \]

The first order term gives the usual expression for the tunneling current, which is described in terms of the single-particle spectral function \[ \Gamma. \] Following the procedure similar to that in Sec.II, we obtain the higher-order contribution

\[
J_\sigma(k, t) = -\frac{1}{\hbar^4} \sum_{\sigma'} \sum_{k'} \sum_{\omega'\eta} |\gamma_{\sigma'}|^2 |\gamma_{\sigma'}'|^2 G_{\sigma\sigma'}(k', k_\sigma, \omega'' - i\eta, \omega_\sigma' - i\eta, \omega_\sigma' + i\eta) \\
-\frac{1}{\hbar^4} \sum_{\sigma'} \sum_{k'} \sum_{\omega'\eta} |\gamma_{\sigma'}|^2 |\gamma_{\sigma'}'|^2 G'_{\sigma\sigma'}(k', k_\sigma, \omega'' - i\eta, \omega_\sigma' - i\eta, \omega_\sigma' + i\eta) \\
-\frac{1}{\hbar^4} \sum_{\sigma'} \sum_{k'} \sum_{\omega'\eta} |\gamma_{\sigma'}|^2 |\gamma_{\sigma'}'|^2 G''_{\sigma\sigma'}(k', k_\sigma, \omega'' - i\eta, \omega_\sigma' - i\eta, \omega_\sigma' + i\eta), \tag{49}\]

where we introduced the three kinds of the correlation functions:

\[
i G_{\sigma\sigma'}(k', k_\sigma, t_1, t_2, t_3) = \langle \hat{T} \left[ \hat{c}_{k_\sigma}^{\dagger}(t_1) \hat{c}_{-k_\sigma}^{\dagger}(t_2) \right] \hat{c}_{-k'\sigma'}(0) \hat{c}_{k'\sigma'}(t_3) \rangle_{t_0} \theta(-t_1)\theta(-t_2)\theta(-t_3), \tag{50}\]

\[
i G'_{\sigma\sigma'}(k', k_\sigma, t_1, t_2, t_3) = -\langle \hat{c}_{k_\sigma}^{\dagger}(t_2) \hat{c}_{k_\sigma}(0) \hat{c}_{k'\sigma'}(t_1) \hat{c}_{k'\sigma'}(t_3) \rangle_{t_0} \theta(t_1 - t_3)\theta(-t_1)\theta(-t_2)\theta(-t_3), \tag{51}\]

\[
i G''_{\sigma\sigma'}(k', k_\sigma, t_1, t_2, t_3) = \theta(-t_1)\theta(-t_2)\theta(-t_3)\theta(t_3 - t_1) \times \left\{ -\theta(t_1 - t_2) \langle \hat{c}_{-k_\sigma}^{\dagger}(t_2) \hat{c}_{k_\sigma}(t_1) \hat{c}_{k'\sigma'}(t_3) \hat{c}_{k_\sigma}(0) \rangle_{t_0} \\
+ \theta(t_3 - t_2)\theta(t_2 - t_1) \langle \hat{c}_{k_\sigma}(t_1) \hat{c}_{k'\sigma'}(t_2) \hat{c}_{k_\sigma}(t_3) \hat{c}_{k_\sigma}(0) \rangle_{t_0} \\
- \theta(t_2 - t_3) \langle \hat{c}_{k_\sigma}(t_1) \hat{c}_{k'\sigma'}(t_3) \hat{c}_{-k_\sigma}(t_2) \hat{c}_{k_\sigma}(0) \rangle_{t_0} \right\}. \tag{52}\]

Using the energy representation described in Sec. III, one can also express the two-particle contribution to the single-particle current function in the Lehmann representation

\[
J_\sigma(\omega) = \frac{1}{\hbar^4} \sum_{\sigma'} \sum_{k'} \sum_{\omega'\eta} |\gamma_{\sigma'}|^2 |\gamma_{\sigma'}'|^2 \sum_{n,m,l,k} \rho_n \left[ -\frac{\langle n|\hat{c}_{-k_\sigma}^{\dagger}|m\rangle\langle m|\hat{c}_{k_\sigma}|l\rangle}{\omega_{1\sigma'} - (E_k - E_n)/\hbar - i\eta} \frac{\langle l|\hat{c}_{k'\sigma'}|k\rangle\langle k|\hat{c}_{k'\sigma'}|n\rangle}{\omega_{2\sigma} + (E_n - E_m)/\hbar - i\eta} \right] \frac{1}{(E_l + E_n - 2E_k)/\hbar - 2i\eta}. \]


contribution to the single-particle current in the unitarity regime. In contrast to the two-particle current, one can only see the two-particle of DPE current, where the weight of condensed pair contribution increases with increasing pairing interaction. We note that this behavior of small peak from contribution of condensed pairs near the BCS-BCS crossover in Fig. 6. In the BEC regime, the contribution of uncorrelated pair states.

One can see that the first terms in the square brackets represent the contribution from the tunneling a condensate pair, while the second terms represent the contribution of uncorrelated pair states.

Using BCS-Leggett’s theory as in Sec. III A, we obtain

\[
J_\uparrow = \frac{2}{\hbar^2} |\gamma_\uparrow|^2 |\gamma_\downarrow|^2 \\
\times \text{Im} \left[ \frac{v^2_{k\uparrow} v^2_{k\uparrow}'}{(\omega_{1\uparrow} + \omega_{2\uparrow} + 2\mu / \hbar - 2i\eta) \delta_{k\uparrow,k'}} + \frac{v^2_{k\downarrow} v^2_{k\downarrow}'}{(\omega_{1\downarrow} + \omega_{2\downarrow} - (E_{k'} + E_{k} - 2\mu) / \hbar - 2i\eta)} \right] \\
\times \left[ \frac{1}{(\omega_{1\uparrow} - (E_{k'} - \mu) / \hbar - i\eta)} \right]^2
\]

One can see that the first terms in the square brackets represent the contribution from the tunneling a condensate pair, while the second terms represent the contribution of uncorrelated pair states.

B. numerical calculations

Figure 6 shows intensity of the two-particle contribution to the single-particle current in the BCS-BCS crossover in Fig. 6. In the BEC regime, the contribution of uncorrelated pair states is much larger than that of condensed pairs. In the BCS side, however, we can see a small peak from contribution of condensed pairs near \(k/k_F = 1\). The weight of condensed pair contribution decreases with increasing pairing interaction. We note that this behavior of condensed pair contribution is very different from that of condensed pair contribution of DPE current, where the weight of condensed pair contribution increases with increasing pairing interaction. In contrast to the two-particle current, one can only see the two-particle contribution to the single-particle current in the unitarity regime.
FIG. 6. (Color online) Intensity of single-particle current. The values of the pairing interaction $(k_Fa_s)^{-1}$ are (a) -1, (b) 0, and (c) 1.

V. CONCLUSION

We provided a general formalism for DPE current from superfluid Fermi gases within the framework of the time-dependent perturbation theory. Using this formalism, we studied DPE current in superfluid Fermi gases in the BCS-BEC crossover at $T = 0$ within the framework of BCS-Leggett’s theory. From the intensity of two-particle spectral densities and energy distributions of DPE current, we can identify the contribution of condensed pairs and uncorrelated states with the energy of peaks. The peak at $\hbar \omega = 0$ is the contribution of
condensed pairs and other peaks are contribution of uncorrelated states. DPE current as a
function of the energy also showed the very different contribution of uncorrelated pair states
and condensed pair components.

We also calculated the angular distributions of DPE current in the BCS-BEC crossover. In the BCS side \((1/k_F a_s = -1)\), we show the double peak of DPE current, which corresponds the particles constructing a pair respectively emitted. While in the BEC side \((1/k_F a_s = 1)\) we can see the single peak, which means the pairs emitted as molecules.

For comparison, we showed the contribution of the two-particle tunneling process to the single-particle current. We found that the contribution of uncorrelated pair states is always much larger than that of condensed pairs. A small peak from contribution of condensed pairs appears near \(k/k_F = 1\) in the BCS side.

In summary, the present study showed the possibility of distinguishing between weakly-bound Cooper pairs and tightly-bound molecules. We hope that these results will simulate in further experiment.

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Appendix A: General formulation of the two-particle current

We derive the third-order expressions \((20)\) and \((21)\) for the correlation functions \(\langle \hat{F}_\sigma \rangle\). According to \((19)\), we need the following expectation value:

\[
\left\langle \mathcal{T} \left[ \hat{V}(t') \hat{V}(t'') \hat{V}(t''') \hat{F}(k_1, k_2, k_3, k_4)(t) \right] \right\rangle_{t_0} = \sum_{\sigma'} \sum_{k'} e^{i \eta t'} e^{i \eta t''} e^{i \eta t'''}
\]

\[
\times \left\langle \mathcal{T} \left[ \left( \gamma_\sigma e^{-i \omega_\sigma t'} \hat{b}_{k+q_\sigma}(t') \hat{c}_{k\sigma}(t') + \text{H.c.} \right) \left( \gamma_{\sigma'} e^{-i \omega_{\sigma'} t''} \hat{\gamma}_{\sigma'} \hat{b}_{k'+q_{\sigma'}}(t'') \hat{c}_{k'\sigma'}(t'') + \text{H.c.} \right) \right. \right.
\]

\[
\times \left( \gamma_{\sigma''} e^{-i \omega_{\sigma''} t'''} \hat{b}_{k''+q_{\sigma''}}(t''') \hat{c}_{k''\sigma''}(t''') + \text{H.c.} \right) \left. \hat{b}_{k_1\uparrow}(t) \hat{b}_{k_2\downarrow}(t) \hat{b}_{k_3\downarrow}(t) \hat{c}_{k_4\uparrow}(t) \right\rangle_{t_0}. \tag{A1}
\]

Collecting the terms that makes non-vanishing contributions, we obtain

\[
\left\langle \mathcal{T} \left[ \hat{V}(t') \hat{V}(t'') \hat{V}(t''') \hat{F}(k_1, k_2, k_3, k_4)(t) \right] \right\rangle_{t_0} = 3 \sum_{\sigma''} \sum_{k''} \sum_{\sigma'} \sum_{k'} \sum_{\sigma} e^{i \eta t'} e^{i \eta t''} e^{i \eta t'''} \gamma_\sigma \gamma_{\sigma'} \gamma_{\sigma''} e^{i \omega_\sigma t'} e^{i \omega_{\sigma'} t''} e^{-i \omega_{\sigma''} t'''}
\]
Using this result in (A3), we obtain

\[
\langle \mathcal{T} \left[ \hat{V}(t') \hat{V}(t'') \hat{V}(t''') \hat{F}(k_1, k_2, k_3, k_4)(t) \right] \rangle_t_0.
\]

Under the assumption that two systems are initially uncoupled, we obtain

\[
3 \sum_{\sigma''} \sum_{k''} \sum_{\sigma'} \sum_{k'} \sum_{\sigma} \sum_{k} e^{i\omega'' t'''} e^{i\omega' t''} e^{i\omega t} e^{-i\omega'' t''} e^{-i\omega' t''} e^{-i\omega t} \times \langle \mathcal{T} [\hat{b}_{k'' \sigma''}(t') \hat{b}_{k' \sigma'}(t'') \hat{b}_{k' \sigma''}(t''') \hat{b}_{k' \sigma'}(t') \hat{b}_{k'' \sigma''}(t'') \hat{b}_{k'' \sigma''}(t''') \hat{b}_{k'' \sigma''}(t') \hat{b}_{k'' \sigma''}(t'')] \rangle_t_0.
\]

Let us now assume that \( \hat{H}_2 \) takes the non-interacting form:

\[
\hat{H}_2 = \sum_{\sigma} \sum_{k} \epsilon_{k \sigma} \hat{b}_{k \sigma} \hat{b}_{k \sigma}^\dagger.
\]

In this case, we can use the Wick’s theorem for the state 2, and use

\[
\hat{b}_{k \sigma}(t) = \hat{b}_{k \sigma} e^{-i\epsilon_{k \sigma} t / \hbar}, \quad \hat{b}_{k \sigma}^\dagger(t) = \hat{b}_{k \sigma} e^{i\epsilon_{k \sigma} t / \hbar},
\]

Furthermore, we assume that there is no particles in the initial state at \( t = t_0 \), and thus use

\[
\langle \hat{b}_{k \sigma} \hat{b}_{k' \sigma'} \rangle_{t_0} = \delta_{\sigma \sigma'} \delta_{k k'}.
\]

With these assumptions, we obtain

\[
\langle \mathcal{T} [\hat{b}_{k'' \sigma''}(t') \hat{b}_{k' \sigma'}(t'') \hat{b}_{k' \sigma''}(t''') \hat{b}_{k'' \sigma''}(t') \hat{b}_{k'' \sigma''}(t'')] \rangle_t_0.
\]

Using this result in (A3), we obtain

\[
\langle \mathcal{T} \left[ \hat{V}(t') \hat{V}(t'') \hat{V}(t''') \hat{F}(k_1, k_2, k_3, k_4)(t) \right] \rangle_t_0.
\]
Here we have made use of the fact that in equilibrium $\Theta(t', t') \Theta(t, t'') e^{i\omega t' t''} e^{-ik_{14}(t'-t)}/h e^{-i\epsilon k_{14}(t''-t)/h} e^{-ie k_{14}(t-t'')}/h$.

Using the above result in Eq. (A1), we obtain

$$
\langle \hat{F}_\uparrow (k_1, k_2, k_3, k_4) \rangle^{(3)}_t = \left( -\frac{i}{\hbar} \right)^3 \gamma^*_\uparrow |\gamma_\downarrow|^2 e^{i\omega t} \int_{t_0}^{t_0} dt' \int_{t_0}^{t} dt'' \int_{t_0}^{t} dt''' e^{i\epsilon t'} e^{i\epsilon t''} e^{i\epsilon t'''} \Theta(t', t) \Theta(t'', t) \Theta(t, t''')
\times e^{i(\omega t - \epsilon k_{14}/h)(t'-t)} e^{i(\omega t - \epsilon k_{14}/h)(t''-t)} e^{-i(\omega t - \epsilon k_{14}/h)(t'''-t)}
\times \langle \hat{T} \left[ c_{k_2 - q_1}^{\dagger}(t') c_{k_3 - q_1}^{\dagger}(t'') c_{k_4}^{\dagger}(t) \right] \rangle_{t_0}. \tag{A8}
$$

Here we recall that the time integral goes from $t_0$ to $t$ on the chronological branch and goes back from $t$ to $t_0$ on the antichronological branch, and thus one always have $t', t'', t''' < t$. On the other hand, in order to make a non-vanishing contribution one must have $\Theta(t', t) \Theta(t'', t) \Theta(t, t''') = 1$. Therefore $t', t''$ should be on the anti-chronological branch and $t'''$ should be on the chronological branch. We can thus project on the contour-path integral to the real time axis as

$$
\langle \hat{F}_\uparrow (k_1, k_2, k_3, k_4) \rangle^{(3)}_t = \left( -\frac{i}{\hbar} \right)^3 \gamma^*_\uparrow |\gamma_\downarrow|^2 e^{i\omega t} \int_{t_0}^{t} dt' \int_{t_0}^{t} dt'' \int_{t_0}^{t} dt''' e^{i\epsilon t'} e^{i\epsilon t''} e^{i\epsilon t'''} \Theta(t, t') \Theta(t'', t) \Theta(t, t''')
\times e^{i(\omega t - \epsilon k_{14}/h)(t'-t)} e^{i(\omega t - \epsilon k_{14}/h)(t''-t)} e^{-i(\omega t - \epsilon k_{14}/h)(t'''-t)}
\times \langle \hat{T} \left[ c_{k_2 - q_1}^{\dagger}(t') c_{k_3 - q_1}^{\dagger}(t'') c_{k_4}^{\dagger}(t) \right] \rangle_{t_0}. \tag{A9}
$$

Introducing the relative time coordinates

$$
t_1 = t' - t, \quad t_2 = t'' - t, \quad t_3 = t''' - t, \tag{A11}
$$

we obtain

$$
\langle \hat{F}_\uparrow (k_1, k_2, k_2, k_1 - q_1) \rangle^{(3)}_t = \left( -\frac{i}{\hbar} \right)^3 \gamma^*_\uparrow |\gamma_\downarrow|^2 e^{i\omega t} \int_{t_0}^{t_1} dt_1 \int_{t_0}^{t_2} dt_2 \int_{t_0}^{t_3} dt_3
e^{i\eta(t_1 + t_2)} e^{i\eta(t_2 + t_3)} e^{i(\omega t - \epsilon k_{14}/h)t_1} e^{i(\omega t - \epsilon k_{14}/h)t_2} e^{i(\omega t - \epsilon k_{14}/h)(t_2 - t_3)}
\times \langle \hat{T} \left[ c_{k_2 - q_1}^{\dagger}(t_2) c_{k_1 - q_1}^{\dagger}(t_1) \right] \rangle_{t_0}. \tag{A12}
$$

Here we have made use of the fact that in equilibrium $\langle \hat{A}(t_1) \hat{B}(t_2) \rangle_{t_0} = \langle \hat{A}(t_1 - t_2) \hat{B}(0) \rangle_{t_0}$ and so on.
Appendix B: Lehmann representation of the two-particle current

Using the energy representation (29), the two-particle correlation functions can be expressed as

\[
iG^\uparrow(\mathbf{k}_1', \mathbf{k}_2', t_1, t_2, t_3) = \Theta(-t_1)\Theta(-t_2)\Theta(-t_3) \sum_n \sum_m \sum_l \sum_k \rho_n
\]

\[
\times \left[ \Theta(t_1 - t_2)e^{i(E_n - E_m)\tau_2/\hbar}e^{i(E_m - E_l)\tau_1/\hbar}e^{i(E_k - E_n)\tau_3/\hbar}
\right.
\]

\[
\times \langle n|\hat{c}_{\mathbf{k}_4 \downarrow}^\dagger|l\rangle \langle l|\hat{c}_{\mathbf{k}_1 \uparrow}^\dagger|k\rangle \langle k|\hat{c}_{\mathbf{k}_2 \downarrow}^\dagger|n\rangle
\]

\[
- \Theta(t_2 - t_1)e^{i(E_n - E_m)\tau_1/\hbar}e^{i(E_m - E_l)\tau_2/\hbar}e^{i(E_k - E_n)\tau_3/\hbar}
\]

\[
\times \langle n|\hat{c}_{\mathbf{k}_1 \uparrow}^\dagger|m\rangle \langle m|\hat{c}_{\mathbf{k}_3 \downarrow}^\dagger|l\rangle \langle l|\hat{c}_{\mathbf{k}_1 \uparrow}^\dagger|k\rangle \langle k|\hat{c}_{\mathbf{k}_2 \downarrow}^\dagger|n\rangle \right].
\]  

(B1)

Using the integral representation of the step function,

\[
\Theta(t) = \lim_{\delta \to 0^+} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{e^{i\omega t}}{\omega - i\delta} = -\lim_{\delta \to \delta^+} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{e^{-i\omega t}}{\omega + i\delta},
\]

and taking Fourier transform of (B1), we obtain

\[
G^\uparrow(\mathbf{k}_1, \mathbf{k}_2, \omega_1, \omega_2)
\]

\[
= \sum_n \sum_m \sum_l \sum_k \rho_n \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{-1}{\omega + i\delta}
\]

\[
\times \left[ \frac{\langle n|\hat{c}_{\mathbf{k}_4 \downarrow}^\dagger|m\rangle \langle m|\hat{c}_{\mathbf{k}_1 \uparrow}^\dagger|l\rangle \langle l|\hat{c}_{\mathbf{k}_1 \uparrow}^\dagger|k\rangle \langle k|\hat{c}_{\mathbf{k}_2 \downarrow}^\dagger|n\rangle}{\omega_1 - \omega + E_m/\hbar - E_l/\hbar - i\eta}
\]

\[
\times \frac{1}{\omega_2 + \omega + E_n/\hbar - E_m/\hbar - i\eta}
\]

\[
\times \frac{1}{E_k/\hbar - E_n/\hbar - \omega_2 - i\eta}
\]

\[
\times \frac{\langle n|\hat{c}_{\mathbf{k}_1 \uparrow}^\dagger|m\rangle \langle m|\hat{c}_{\mathbf{k}_3 \downarrow}^\dagger|l\rangle \langle l|\hat{c}_{\mathbf{k}_1 \uparrow}^\dagger|k\rangle \langle k|\hat{c}_{\mathbf{k}_2 \downarrow}^\dagger|n\rangle}{\omega_1 + \omega + E_n/\hbar - E_m/\hbar - i\eta}
\]

\[
\times \frac{1}{\omega_2 - \omega + E_m/\hbar - E_l/\hbar - i\eta}
\]

\[
\times \frac{1}{E_k/\hbar - E_n/\hbar - \omega_2 - i\eta}
\right].
\]  

(B3)

Taking the limit \(\delta \to 0^+\) is implied in the above expression. Carrying out the integral over \(\omega\), we obtain

\[
G^\uparrow(\mathbf{k}_1, \mathbf{k}_2, \omega_1, \omega_2) = -\sum_n \sum_l \rho_n
\]

20
With the Hamiltonian given by (1), we have
\[
\times \left\{ \frac{1}{\omega_1 + \omega_2 - (E_l - E_n)/h - 2i\eta} \sum_m \frac{\langle l|\hat{c}_{k\downarrow}^\dagger|m\rangle\langle m|\hat{c}_{k\uparrow}^\dagger|n\rangle}{\omega_2 - (E_m - E_n)/h + i\eta} \right\}
- \left[ \frac{1}{\omega_2 + \omega_1 - (E_l - E_n)/h - 2i\eta} \sum_m \frac{\langle l|\hat{c}_{k\uparrow}^\dagger|m\rangle\langle m|\hat{c}_{k\downarrow}^\dagger|n\rangle}{\omega_1 - (E_m - E_n)/h + i\eta} \right]^*
\times \left[ \sum_m \frac{\langle l|\hat{c}_{k\downarrow}^\dagger|m\rangle\langle m|\hat{c}_{k\uparrow}^\dagger|n\rangle}{\omega_2 - (E_m - E_n)/h + i\eta} \right].
\]

Similarly, we obtain
\[
G_\downarrow(k_1, k_2, \omega_1, \omega_2) = -\sum_n \sum_l \rho_n \left\{ \frac{1}{\omega_1 + \omega_2 - (E_l - E_n)/h - 2i\eta} \sum_m \frac{\langle l|\hat{c}_{k\downarrow}^\dagger|m\rangle\langle m|\hat{c}_{k\uparrow}^\dagger|n\rangle}{\omega_2 - (E_m - E_n)/h + i\eta} \right\}
- \left[ \frac{1}{\omega_2 + \omega_1 - (E_l - E_n)/h - 2i\eta} \sum_m \frac{\langle l|\hat{c}_{k\uparrow}^\dagger|m\rangle\langle m|\hat{c}_{k\downarrow}^\dagger|n\rangle}{\omega_1 - (E_m - E_n)/h + i\eta} \right]^*
\times \left[ \sum_m \frac{\langle l|\hat{c}_{k\downarrow}^\dagger|m\rangle\langle m|\hat{c}_{k\uparrow}^\dagger|n\rangle}{\omega_2 - (E_m - E_n)/h + i\eta} \right].
\]

Appendix C: Single-particle current

The single-particle density in the scattered state is given by
\[
n_\sigma(k, t) = \langle \hat{b}_{k\sigma}^\dagger \hat{b}_{k\sigma} \rangle_t. \tag{C1}
\]

Then, the single-particle current is given by
\[
J_\sigma(k, t) = \frac{d}{dt} n_\sigma(k, t) = \frac{i}{\hbar} \langle [\hat{b}_{k\sigma}^\dagger \hat{b}_{k\sigma}, \hat{H}] \rangle_t. \tag{C2}
\]

With the Hamiltonian given by (1), we have
\[
J_\sigma(k, t) = \frac{i}{\hbar} e^{\eta t} \left( \gamma_\sigma e^{-i\omega_\sigma t} \langle \hat{b}_{k\sigma}^\dagger \hat{c}_{k-q\sigma} \rangle_t - \gamma_\sigma^* e^{i\omega_\sigma t} \langle \hat{c}_{k-q\sigma}^\dagger \hat{b}_{k\sigma} \rangle_t \right). \tag{C3}
\]

Employing the time-dependent perturbation theory, we can express the expectation values of \( b^\dagger c \) and \( c^\dagger b \) as
\[
\langle b^\dagger c \rangle = \langle b^\dagger c \rangle^{(1)} + \langle b^\dagger c \rangle^{(3)} \tag{C4}
\]
\[
\langle c^\dagger b \rangle = \langle c^\dagger b \rangle^{(1)} + \langle c^\dagger b \rangle^{(3)} \tag{C5}
\]
where the superscript \((n)\) denotes the \(n\)-th order contribution. The first order term gives the usual expression for the tunneling current, which is described in terms of the single-particle spectral function. We are now interested in the third-order contributions

\[
\langle \hat{b}_{\mathbf{k}, \sigma} \hat{c}_{\mathbf{q}, \sigma} \rangle^{(3)}_t = \frac{1}{6} \left( -\frac{i}{\hbar} \right)^3 \int \frac{dt'}{c} \int \frac{dt''}{c} \int \frac{dt'''}{c} \langle T \left[ \hat{V}(t') \hat{V}(t'') \hat{V}(t''') \hat{b}_{\mathbf{k}, \sigma}(t) \hat{c}_{\mathbf{q}, \sigma}(t) \right] \rangle_{t_0}, \tag{C6}
\]

\[
\langle \hat{c}_{\mathbf{k}, \sigma} \hat{b}_{\mathbf{q}, \sigma} \rangle^{(3)}_t = \frac{1}{6} \left( -\frac{i}{\hbar} \right)^3 \int \frac{dt'}{c} \int \frac{dt''}{c} \int \frac{dt'''}{c} \langle T \left[ \hat{V}(t') \hat{V}(t'') \hat{V}(t''') \hat{b}_{\mathbf{k}, \sigma}(t) \hat{c}_{\mathbf{q}, \sigma}(t) \right] \rangle_{t_0}. \tag{C7}
\]

The non-vanishing contribution is

\[
\langle \hat{b}_{\mathbf{k}, \sigma} \hat{c}_{\mathbf{q}, \sigma} \rangle^{(3)}_t = \frac{i}{2\hbar} \sum_{\sigma, \sigma', \sigma''} \gamma^\sigma_{\sigma'} \gamma^\sigma_{\sigma''} \int \frac{dt'}{c} \int \frac{dt''}{c} \int \frac{dt'''}{c} \langle T \left[ \hat{V}(t') \hat{V}(t'') \hat{V}(t''') \hat{b}_{\mathbf{k} + \mathbf{q}, \sigma}(t') \hat{c}_{\mathbf{k}', \sigma'}(t'') \hat{c}_{\mathbf{q}, \sigma''}(t''') \right] \hat{b}_{\mathbf{k}, \sigma}(t) \hat{c}_{\mathbf{q}, \sigma}(t) \rangle_{t_0}, \tag{C8}
\]

With the assumption that the two internal states are initially uncoupled and the scattered state is a free gas, the above correlation function can be decoupled as

\[
\langle T \left[ \hat{c}_{\mathbf{k}', \sigma'}(t') \hat{b}_{\mathbf{k} + \mathbf{q}, \sigma}(t') \hat{c}_{\mathbf{k}'', \sigma''}(t'') \hat{b}_{\mathbf{k}', \sigma'}(t'') \hat{c}_{\mathbf{k}, \sigma}(t) \hat{c}_{\mathbf{q}, \sigma}(t) \right] \rangle_{t_0}
= \langle T \left[ \hat{c}_{\mathbf{k}', \sigma'}(t') \hat{c}_{\mathbf{k}'', \sigma''}(t'') \hat{b}_{\mathbf{k}', \sigma'}(t'') \hat{c}_{\mathbf{k}, \sigma}(t) \hat{c}_{\mathbf{q}, \sigma}(t) \right] \rangle_{t_0}
\times \langle T \left[ \hat{b}_{\mathbf{k} + \mathbf{q}, \sigma}(t') \hat{c}_{\mathbf{k}', \sigma'}(t'') \hat{c}_{\mathbf{q}, \sigma''}(t'') \hat{b}_{\mathbf{k}, \sigma}(t) \right] \rangle_{t_0}
= -\Theta(t', t''') \Theta(t'', t') \delta_{\mathbf{k}', \mathbf{k}} \delta_{\sigma', \sigma} \delta_{\sigma''} \delta_{\mathbf{q}, \mathbf{q}} \delta_{\sigma''} \delta_{\sigma} e^{-i\epsilon_{\mathbf{k}, \sigma}(t'-t)} e^{-i\epsilon_{\mathbf{q}, \sigma''}(t''-t')} \tag{C9}
\]

Using (C8) and (C9), we obtain

\[
\langle \hat{b}_{\mathbf{k}, \sigma} \hat{c}_{\mathbf{q}, \sigma} \rangle^{(3)}_t
= \frac{i}{\hbar} \sum_{\sigma'} \gamma^\sigma_{\sigma'} |\gamma_{\sigma'}|^2 \int \frac{dt'}{c} \int \frac{dt''}{c} \Theta(t', t') \Theta(t', t''') \Theta(t', t'') e^{-i\epsilon_{\mathbf{k}, \sigma}(t'-t)/\hbar} e^{-i\epsilon_{\mathbf{q}, \sigma'}(t''-t')/\hbar}
\times \langle T \left[ \hat{c}_{\mathbf{k}, \sigma} \hat{c}_{\mathbf{q}, \sigma} \right] \rangle_{t_0}. \tag{C10}
\]

In order to express (C10) in terms of the real-time integral, we split the contour integral into the forward (\(\mathcal{C}^+\)) and return (\(\mathcal{C}^-\)) paths. Then the contour integral involving the product of step functions can be written as

\[
\int \frac{dt'}{c} \int \frac{dt''}{c} \int \frac{dt'''}{c} \Theta(t', t') \Theta(t', t''')
= \int_{\mathcal{C}^+} \frac{dt''}{c} \int \frac{dt'}{c} \int \frac{dt'''}{c} \Theta(t' - t''')
+ \int_{\mathcal{C}^-} \frac{dt'}{c} \int \frac{dt''}{c} \int \frac{dt'''}{c} \Theta(t'' - t') \tag{C11}
\]
Figure 6 depicts the three contributions in the square bracket of (C). We now calculate the contribution depicted by Fig. 7 (b), which is given by

\[
\int_{t_0} dt' \int_{t_0} dt'' e^{i(\omega_0 + \eta) t'} e^{i(\omega_2 - \eta) t''} e^{-i \epsilon_{k_\alpha} t''} e^{i \epsilon_{k_\nu} t''} / \hbar e^{-i \epsilon_{k_\sigma} t''} / \hbar \\
\langle T \left[ \hat{c}_{k-\sigma} \sigma \left( t'' \right) \hat{c}_{k+\sigma} \sigma \left( t' \right) \hat{c}_{k+\sigma} \sigma \left( t'' \right) \hat{c}_{k-\sigma} \sigma \left( t \right) \right] \rangle / \hbar \\
= e^{i(\omega_0 - \eta) t} \int_{t_0} dt_1 \int_{t_0} dt_2 \int_{t_0} dt_3 e^{i(\omega_{\sigma'} - \epsilon_{k_\sigma} - \epsilon_{k_\nu}) / \hbar} e^{i(\omega_0 - \epsilon_{k_\sigma}) / \hbar} e^{i(\omega_2 - \epsilon_{k_\nu}) / \hbar} e^{i(\omega_2 - \epsilon_{k_\sigma}) / \hbar + \eta) t_3} \\
\langle \bar{T} \left[ \hat{c}_{k+\sigma} \sigma \left( t_1 \right) \hat{c}_{k-\sigma} \sigma \left( t_2 \right) \right] \hat{c}_{k-\sigma} \sigma \left( 0 \right) \hat{c}_{k+\sigma} \sigma \left( t_3 \right) \rangle / \hbar.
\]

(C12)

Here we have introduced the relative time coordinates

\[
t_1 = t' - t, \quad t_2 = t'' - t, \quad t_3 = t''' - t.
\]

(C13)

Using the notations \( k - \sigma = k_\sigma, \omega_{\sigma'} = \omega_\sigma - \epsilon_{k_\nu} \), \( \omega'_{\sigma'} = \omega_{\sigma' - \epsilon_{k_\sigma}} \), we obtain the expression for the contribution Fig. 7 (b) to the single-particle current as

\[
J_{(b)}^\sigma = -\frac{1}{\hbar^4} \sum_{\sigma'} \sum_{k'} |\gamma_{\sigma'}|^2 |\gamma_{\sigma'}|^2 G_{\sigma\sigma'}(k', k_\sigma, \omega_{\sigma'} - i \eta, \omega_{\sigma'} - i \eta, \omega_{\sigma'} + i \eta),
\]

(C14)

where we defined the Fourier transforms by

\[
G_{\sigma\sigma'}(k_1, k_2, \omega_{1\sigma'}, \omega_{2\sigma}, \omega_{3\sigma'}) = \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \int_{-\infty}^{\infty} dt_3 e^{i \omega_{1\sigma'} t_1} e^{i \omega_{2\sigma} t_2} e^{-i \omega_{3\sigma} t_3} G_{\sigma\sigma'}(k_1, k_2, t_1, t_2, t_3),
\]

(C15)

with

\[
iG_{\sigma\sigma'}(k', k_\sigma, t_1, t_2, t_3) = \langle \bar{T} \left[ \hat{c}_{k+\sigma} \sigma \left( t_1 \right) \hat{c}_{k-\sigma} \sigma \left( t_2 \right) \right] \hat{c}_{k-\sigma} \sigma \left( 0 \right) \hat{c}_{k+\sigma} \sigma \left( t_3 \right) \rangle / \hbar \theta(-t_1) \theta(-t_2) \theta(-t_3)
\]

(C16)
The contribution of Fig. 7 (a) is given by

\[
\int_\mathcal{C} dt'' \int_\mathcal{C} dt' \int_\mathcal{C} dt''' \theta(t'' - t''') e^{i(\omega - \omega_0) t''} e^{i(\omega_0 - \omega_0) (t' - t''')} e^{-i\epsilon_{k_0} (t'' - t)} e^{-i\epsilon_{k_0 + q_{\sigma'}} (t' - t'')} e^{-i\epsilon_{k_0 + q_{\sigma'}} (t' - t''')} \\
\langle T \left[ \hat{c}_{k - q_{\sigma} \sigma} (t'') \hat{c}^\dagger_{k' \sigma'} (t') \hat{c}_{k' \sigma'} (t''') \hat{c}_{k - q_{\sigma} \sigma} (t) \right] \rangle_t_0 \\
= -e^{i(\omega - \omega_0) t} \int_0^t dt_1 \int_0^t dt_2 \int_0^t dt_3 \theta(t_1 - t_3) \\
\times e^{i(\omega_0 - \epsilon_{k_0 + q_{\sigma}}) (t_1)} e^{i(\omega - \epsilon_{k_0 + q_{\sigma}}) (t_2)} e^{-i(\omega_0 - \epsilon_{k_0 + q_{\sigma}}) (t_3)} \\
\times e^{-i(\omega_0 - \epsilon_{k_0 + q_{\sigma}}) (t_4)} (\hat{c}_{k - q_{\sigma} \sigma} (t_2) \hat{c}_{k - q_{\sigma} \sigma} (0) \hat{c}_{k' \sigma'} (t_1) \hat{c}_{k' \sigma'} (t_3))_t_0. \\
\text{(C17)}
\]

Therefore we obtain its contribution to the single-particle current as

\[
J^{(a)}_\sigma = -\frac{1}{\hbar^4} \sum_{\sigma'} \sum_{k'} |\gamma_{\sigma'}|^2 |\gamma_{\sigma}|^2 G_{\sigma \sigma'} (k', k, \omega, -i\eta, \omega' - i\eta, \omega'' + i\eta), \\
\text{(C18)}
\]

where

\[
G_{\sigma \sigma'} (k_1, k_2, \omega_{1 \sigma'}, \omega_{2 \sigma}, \omega_{3 \sigma}) = \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \int_{-\infty}^{\infty} dt_3 e^{i\omega_{1 \sigma'} t_1} e^{i\omega_{2 \sigma} t_2} e^{-i\omega_{3 \sigma} t_3} G_{\sigma \sigma'} (k_1, k_2, t_1, t_2, t_3), \\
\text{(C19)}
\]

and the correlation function is given by

\[
iG_{\sigma \sigma'} (k', k, t_1, t_2, t_3) = -\langle \hat{c}_{k \sigma} (t_2) \hat{c}_{k \sigma} (0) \hat{c}_{k' \sigma'} (t_1) \hat{c}_{k' \sigma'} (t_3) \rangle_t_0 \theta(t_1 - t_3) \theta(-t_1) \theta(-t_2) \theta(-t_3). \\
\text{(C20)}
\]

The contribution of Fig. 7 (c) is given by

\[
\int_\mathcal{C} dt'' \int_\mathcal{C} dt' \int_\mathcal{C} dt''' \theta(t'' - t') e^{i(\omega - \omega_0) t''} e^{i(\omega_0 - \omega_0) (t' - t''')} e^{-i\epsilon_{k_0} (t' - t)} e^{-i\epsilon_{k_0 + q_{\sigma'}} (t' - t''')} e^{-i\epsilon_{k_0 + q_{\sigma'}} (t' - t''')} \\
\langle T \left[ \hat{c}_{k - q_{\sigma} \sigma} (t'') \hat{c}^\dagger_{k' \sigma'} (t') \hat{c}_{k' \sigma'} (t''') \hat{c}_{k - q_{\sigma} \sigma} (t) \right] \rangle_t_0 \\
= -e^{i(\omega - \omega_0) t} \int_0^t dt_1 \int_0^t dt_2 \int_0^t dt_3 e^{i(\omega_0 - \epsilon_{k_0 + q_{\sigma}}) (t_1)} e^{i(\omega - \epsilon_{k_0 + q_{\sigma}}) (t_2)} e^{-i(\omega_0 - \epsilon_{k_0 + q_{\sigma}}) (t_3)} \\
\times \theta(t_3 - t_1) \left\{ -\theta(t_1 - t_2) \langle \hat{c}_{k - q_{\sigma} \sigma} (t_2) \hat{c}^\dagger_{k' \sigma'} (t_1) \hat{c}_{k' \sigma'} (t_3) \hat{c}_{k - q_{\sigma} \sigma} (0) \rangle_t_0 \\
+ \theta(t_3 - t_2) \theta(t_2 - t_1) \langle \hat{c}_{k' \sigma'} (t_1) \hat{c}_{k' \sigma'} (t_3) \hat{c}_{k - q_{\sigma} \sigma} (t_2) \hat{c}_{k - q_{\sigma} \sigma} (0) \rangle_t_0 \\
- \theta(t_2 - t_3) \langle \hat{c}_{k' \sigma'} (t_1) \hat{c}_{k' \sigma'} (t_3) \hat{c}_{k - q_{\sigma} \sigma} (t_2) \hat{c}_{k - q_{\sigma} \sigma} (0) \rangle_t_0 \right\}. \\
\text{(C21)}
\]

Therefore, we obtain its contribution to the single-particle current as

\[
J^{(c)}_\sigma = -\frac{1}{\hbar^4} \sum_{\sigma'} \sum_{k'} |\gamma_{\sigma'}|^2 |\gamma_{\sigma}|^2 G_{\sigma \sigma'} (k', k, \omega, -i\eta, \omega' - i\eta, \omega'' + i\eta), \\
\text{(C22)}
\]

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where we defined
\[
G''_{\sigma'\sigma'}(k_1, k_2, \omega_{1\sigma'}, \omega_{2\sigma}, \omega_{3}) = \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \int_{-\infty}^{\infty} dt_3 e^{i\omega_{1\sigma'} t_1} e^{i\omega_{2\sigma} t_2} e^{-i\omega_{3} t_3} G_{\sigma'\sigma'}''(k_1, k_2, t_1, t_2, t_3),
\] (C23)
with
\[
iG_{\sigma'\sigma'}''(k', k_\sigma, t_1, t_2, t_3)
= \theta(-t_1)\theta(-t_2)\theta(-t_3)\theta(t_3 - t_1) \left\{ -\theta(t_1 - t_2)\langle \hat{c}^\dagger_{k - q_{\sigma \sigma}}(t_2)\hat{c}_{k'\sigma'}(t_1)\hat{c}_{k'\sigma'}(t_3)\hat{c}_{k - q_{\sigma \sigma}}(0) \rangle_{t_0} \\
+ \theta(t_3 - t_2)\theta(t_2 - t_1)\langle \hat{c}^\dagger_{k'\sigma'}(t_1)\hat{c}_{k'\sigma'}(t_2)\hat{c}_{k - k_{\sigma \sigma}}(t_3)\hat{c}_{k - q_{\sigma \sigma}}(0) \rangle_{t_0} \\
- \theta(t_2 - t_3)\langle \hat{c}^\dagger_{k'\sigma'}(t_1)\hat{c}_{k'\sigma'}(t_3)\hat{c}_{k - k_{\sigma \sigma}}(t_2)\hat{c}_{k - q_{\sigma \sigma}}(0) \rangle_{t_0} \right\}.
\] (C24)

Collecting the above results, we obtain the expression for the third-order contribution to the singleparticle current as
\[
J_{\sigma}(k, t) = J^{(a)}_{\sigma} + J^{(b)}_{\sigma} + J^{(c)}_{\sigma}
= -\frac{1}{\hbar^4} \sum_{\sigma'} \sum_{k'} |\gamma_{\sigma'}|^2 |\gamma_{\sigma}|^2 G_{\sigma'\sigma'}(k', k_{\sigma}, \omega_{\sigma'}, -i\eta, \omega_{\sigma'} - i\eta, \omega_{\sigma'} + i\eta)
-\frac{1}{\hbar^4} \sum_{\sigma'} \sum_{k'} |\gamma_{\sigma'}|^2 |\gamma_{\sigma}|^2 G_{\sigma'\sigma}(k', k_{\sigma}, \omega_{\sigma'}, -i\eta, \omega_{\sigma'} - i\eta, \omega_{\sigma'} + i\eta)
-\frac{1}{\hbar^4} \sum_{\sigma'} \sum_{k'} |\gamma_{\sigma'}|^2 |\gamma_{\sigma}|^2 G_{\sigma'\sigma'}''(k', k_{\sigma}, \omega_{\sigma'}, -i\eta, \omega_{\sigma'} - i\eta, \omega_{\sigma'} + i\eta).
\] (C25)

Using the energy representation introduced in Sec III, we can express the two-particle correction function. Taking the limit \(\delta \rightarrow 0^+\), we thus have
\[
G_{\sigma'\sigma'}(k', k - q_{\sigma}, \omega_{1\sigma'}, \omega_{2\sigma}, \omega_{1\sigma})
= \sum_{n, m, l, k} \rho_n \int \frac{d\omega}{2\pi} \frac{1}{\omega + i\delta} \int_{-\infty}^{0} dt_1 \int_{-\infty}^{0} dt_2 \int_{-\infty}^{0} dt_3 e^{i(\omega_{1\sigma'} + \omega_{\sigma}) t_1} e^{i(\omega_{2\sigma} - i\eta) t_2} e^{-i(\omega_{1\sigma'} - \omega_{\sigma}) t_3}
\left[ e^{-i\omega(t_2 - t_1)} e^{i(E_n - E_m)t_2/h} e^{i(E_m - E_l)t_1/h} e^{i(E_k - E_m)t_3/h} \langle n|\hat{c}_{k'\sigma'}|m\rangle \langle m|\hat{c}_{k - q_{\sigma \sigma}}^\dagger |l\rangle \langle l|\hat{c}_{k - q_{\sigma \sigma}}|k\rangle \langle k|\hat{c}_{k'\sigma'}|n\rangle \\
- e^{-i\omega(t_1 - t_2)} e^{i(E_n - E_m)t_1/h} e^{i(E_m - E_l)t_2/h} e^{i(E_k - E_m)t_3/h} \langle n|\hat{c}_{k - q_{\sigma \sigma}}^\dagger |m\rangle \langle m|\hat{c}_{k'\sigma'}^\dagger |l\rangle \langle l|\hat{c}_{k - q_{\sigma \sigma}}|k\rangle \langle k|\hat{c}_{k'\sigma'}|n\rangle \right]
= -\sum_{n, l} \rho_n \int \frac{1}{\omega_{1\sigma'} + \omega_{2\sigma} + (E_n - E_l)/\hbar + 2i\eta} \left[ \sum_{m} \frac{|\langle l|\hat{c}_{k - q_{\sigma \sigma}}|m\rangle \langle m|\hat{c}_{k'\sigma'}|n\rangle|^2}{\omega_{1\sigma'} + (E_n - E_m)/\hbar + i\eta} \\
- \sum_{m} \frac{|\langle l|\hat{c}_{k - q_{\sigma \sigma}}|l\rangle|^2}{\omega_{2\sigma} + (E_m - E_l)/\hbar + i\eta} \right] \left[ \sum_{m} \frac{|\langle l|\hat{c}_{k - q_{\sigma \sigma}}|m\rangle \langle m|\hat{c}_{k'\sigma'}|n\rangle|^2}{\omega_{1\sigma'} + (E_n - E_m)/\hbar + i\eta} \\
- \sum_{m} \frac{|\langle l|\hat{c}_{k - q_{\sigma \sigma}}|l\rangle|^2}{\omega_{2\sigma} + (E_m - E_l)/\hbar + i\eta} \right] \right].
\] (C26)

Similarly, we obtain
\[
G_{\sigma'\sigma'}'(k', k - q_{\sigma}, \omega_{1\sigma'}, \omega_{2\sigma}, \omega_{1\sigma})
\]
Using the above results, we can obtain Eq. (54).

\[ \langle \sigma | \hat{c}_{k-q,\sigma}^\dagger | m \rangle \langle m | \hat{c}_{k-q,\sigma}^\dagger | l \rangle \langle l | \hat{c}_{k',\sigma'}^\dagger | k \rangle \langle k | \hat{c}_{k',\sigma'} | n \rangle \]
\[ \frac{\rho_n}{\omega_{1\sigma'} - (E_k - E_n)/h - i\eta} \]
\[ \frac{1}{\omega_{2\sigma} + (E_n - E_m)/h - i\eta (E_l + E_n - 2E_k)/h - 2i\eta}. \]

\[ (C27) \]

and

\[ G''_{\sigma\sigma'}(k', k - q, \omega_{1\sigma'}, \omega_{2\sigma}, \omega_{1\sigma'}) \]
\[ = \sum_{n,m,l,k} \rho_n \left\{ \frac{\langle n | \hat{c}_{k-q,\sigma}^\dagger | m \rangle \langle m | \hat{c}_{k-q,\sigma}^\dagger | l \rangle \langle l | \hat{c}_{k',\sigma'}^\dagger | k \rangle \langle k | \hat{c}_{k',\sigma'} | n \rangle}{\omega_{1\sigma'} - (E_l - E_k)/h - i\eta \omega_{2\sigma} + (E_n - E_k)/h - i\eta (E_k - E_m)/h} \right. \]
\[ + \frac{\langle n | \hat{c}_{k',\sigma'}^\dagger | m \rangle \langle m | \hat{c}_{k,\sigma}^\dagger | l \rangle \langle l | \hat{c}_{k-q,\sigma}^\dagger | k \rangle \langle k | \hat{c}_{k-q,\sigma} | n \rangle}{\omega_{1\sigma'} + (E_n - E_m)/h - i\eta \omega_{2\sigma} + (E_m - E_l)/h - i\eta \omega_{1\sigma'} - 2\omega_{2\sigma} + (E_k - E_m)/h - i\eta} \]
\[ + \left. \frac{\langle n | \hat{c}_{k',\sigma'}^\dagger | m \rangle \langle m | \hat{c}_{k,\sigma}^\dagger | l \rangle \langle l | \hat{c}_{k-q,\sigma}^\dagger | k \rangle \langle k | \hat{c}_{k-q,\sigma} | n \rangle}{-\omega_{1\sigma'} - (E_n - E_m)/h + i\eta - \omega_{2\sigma} - (E_k - E_l)/h + i\eta - \omega_{1\sigma'} + 2\omega_{2\sigma} + (E_m - E_l)/h} \right\}. \]

\[ (C28) \]

Using the above results, we can obtain Eq. [54].

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