Fermi’s golden rule in a mesoscopic metal ring

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(June 1, 1999)

Abstract

We examine the time-dependent non-equilibrium current in a mesoscopic metal ring threaded by a static magnetic flux $\phi$ that is generated by a time-dependent electric field oscillating with frequency $\omega$. We show that in quadratic order in the field there are three fundamentally different contributions to the current. (a) A time-independent contribution which can be obtained from a thermodynamic derivative. (b) A term increasing linearly in time that can be understood in terms of Fermi’s golden rule. The derivation of this term requires a careful treatment of the infinitesimal imaginary parts that are added to the real frequency $\omega$ when the electric field is adiabatically switched on. (c) Finally, there is also a time-dependent current oscillating with frequency $2\omega$. We suggest an experiment to test our results.

Keywords: persistent currents, non-linear response

PACS numbers: 73.50.Bk, 72.10.Bg, 72.15.Rn
I. INTRODUCTION

Consider a mesoscopic metal ring threaded by a time-dependent magnetic flux \( \phi(t) \) that has a static component \( \phi \) and a part that oscillates with frequency \( \omega \),

\[
\phi(t) = \phi + \phi_\omega \sin(\omega t) .
\]  

(1)

By Faraday’s law of induction, the oscillating part generates a time-dependent electric field directed along the circumference of the ring, \( E(t) = E_\omega \cos(\omega t) \), with amplitude

\[
e LE_\omega = 2\pi\omega \frac{\phi_\omega}{\phi_0} .
\]  

(2)

Here \( L \) is the circumference of the ring, \(-e\) is the charge of the electron, and \( \phi_0 \) is the flux quantum. We would like to know the induced current around the ring. In the limit \( \omega \to 0 \) this is just the usual persistent current \([1,2]\). But what happens for frequencies in the range between \( 10^8 \) and \( 10^{13} \) Hz, which for experimentally relevant rings \([3,4]\) corresponds to \( \Delta \ll \omega \ll \tau^{-1} \)? Here \( \Delta \) is the average level spacing at the Fermi energy, and \( \tau \) is the elastic lifetime. We use units where \( \hbar \) is set equal to unity. This problem has first been studied by Kravtsov and Yudson \([5]\) (KY), who found that in quadratic order the time-dependent field induces (among other terms that oscillate) a time-independent non-equilibrium current \( I_0^{(2)} \).

Calculating the disorder average of this current perturbatively, KY found that it has the peculiar property that for frequencies exceeding the Thouless energy \( E_c = \hbar D/L^2 \) (where \( D \) is the diffusion coefficient) the average of \( I_0^{(2)} \) does not vanish exponentially, but only as \( \omega^{-2} \). This is in disagreement with the intuitive expectation that the external frequency \( \omega \) leads to a similar exponential suppression of this mesoscopic non-equilibrium current as a dephasing rate in the case of the equilibrium persistent current \([3,4]\). The perturbative calculation of KY is based on the assumption of a continuous energy-spectrum, which means that the level-broadening due to dephasing, \( 1/\tau_\varphi \), must exceed the average level spacing at the Fermi energy, \( \Delta \). If we assume that for low temperature \( T \) the dominant dephasing effect comes from electron-electron interactions, a simple estimate \([8]\) shows that \( 1/\tau_\varphi(\omega) < \Delta \) for \( |\omega| \leq E_c \) in the limit \( T \to 0 \). Hence, for frequencies smaller than the Thouless energy the spectrum is discrete and the perturbative analysis breaks down. In this work we shall show that in this case the term considered by KY is not constant, but grows linearly in time, a result which can be understood simply in terms of Fermi’s golden rule of time-dependent perturbation theory.

It is important to point out the difference between the current considered here and the direct current due to the usual photovoltaic effect. It is well known \([4]\) that irradiation of a medium without an inversion center by an alternating electric field can give rise to a direct current (photovoltaic effect). The lack of inversion symmetry can be due to impurities and defects in a finite sample. For mesoscopic junctions the photovoltaic direct current has been studied in Ref. \([10]\). In this case the average current vanishes, because disorder averaging restores the inversion symmetry. In our case, however, we calculate the direct current induced in a mesoscopic ring threaded by a magnetic flux. Because the magnetic flux breaks the time-reversal symmetry, the direct current considered here has a finite disorder average. Thus, the physical origin of a mesoscopic non-equilibrium current discussed in this work is quite different from Ref. \([11]\).
II. THE QUADRATIC RESPONSE FUNCTION: WHAT IS WRONG WITH THE GREEN’S FUNCTION APPROACH?

We consider non-interacting disordered electrons of mass $m$ on a mesoscopic metal ring threaded by the time-dependent magnetic flux given in Eq.(1). Suppose that we have diagonalized the Hamiltonian in the absence of the oscillating flux (i.e. for $\phi_\omega = 0$ in Eq.(1)) for the given realization of the disorder. The time-independent part of the Hamiltonian is then $\hat{H}_0 = \sum_\alpha \varepsilon_\alpha c_\alpha ^\dagger c_\alpha$, where $\varepsilon_\alpha$ are the exact electronic eigen-energies for fixed disorder, which are labeled by appropriate quantum numbers $\alpha$. The operators $c_\alpha ^\dagger$ create electrons in the corresponding eigenstates $|\alpha\rangle$. If we now switch on the time-dependent part of the field, the Hamiltonian becomes $\hat{H} = \hat{H}_0 + \hat{V}(t)$, with

$$\hat{V}(t) = \frac{2\pi}{mL} \delta \phi(t) \sum_{\alpha,\beta} \langle \alpha | \hat{P}_x | \beta \rangle c_\alpha ^\dagger c_\beta$$

$$+ \frac{1}{2m} \left( \frac{2\pi}{L} \delta \phi(t) \right)^2 \sum_\alpha c_\alpha ^\dagger c_\alpha .$$

(3)

Here $\delta \phi(t) = (\phi_\omega/\phi_0) \sin(\omega t)$, and $\hat{P}_x = -id/dx + (2\pi/L)(\phi/\phi_0)$ is the $x$-component of the one particle momentum operator. As usual, the coordinate along the circumference is called the $x$-direction, and we impose periodic boundary conditions. Using standard non-equilibrium Green’s function methods, the contribution to the non-equilibrium current that is quadratic in the external field is easily obtained [11]:

$$I^{(2)}(t) = \frac{(-e)(2\pi)^2}{(mL)^3} \int_0^\infty d\omega_1 d\omega_2 \delta\varphi_{\omega_1} \delta\varphi_{\omega_2}$$

$$\times K^{(2)}(\omega_1, \omega_2) e^{-i(\omega_1+\omega_2)t} ,$$

(4)

where $\varphi_\omega$ is the Fourier transform of the time-dependent part of the flux (i) in units of the flux quantum (i.e. $\phi(t) - \phi = \phi_0 \int d\omega' \delta\varphi_{\omega'} e^{-i\omega't}$) and the response function $K^{(2)}(\omega_1, \omega_2)$ is given by

$$K^{(2)}(\omega_1, \omega_2) = \sum_{\alpha,\beta,\gamma} \frac{P_{\alpha\beta\gamma}}{\varepsilon_\gamma - \varepsilon_\alpha + \omega_1 + \omega_2 + i0}$$

$$\times \left[ \frac{f(\varepsilon_\gamma) - f(\varepsilon_\beta)}{\varepsilon_\gamma - \varepsilon_\beta + \omega_2 + i0} - \frac{f(\varepsilon_\beta) - f(\varepsilon_\alpha)}{\varepsilon_\beta - \varepsilon_\alpha + \omega_1 + i0} \right] ,$$

(5)

with

$$P_{\alpha\beta\gamma} = \langle \alpha | \hat{P}_x | \beta \rangle \langle \beta | \hat{P}_x | \gamma \rangle \langle \gamma | \hat{P}_x | \alpha \rangle .$$

(6)

Here $f(\varepsilon_\alpha) = \langle c_\alpha ^\dagger c_\alpha \rangle$ is the occupation number, which in a grand-canonical ensemble is the Fermi function. Keeping in mind that the time-dependent part of the flux (i) corresponds to

$$\delta\varphi_{\omega'} = \frac{\phi_\omega}{2i\phi_0} [\delta(\omega' + \omega) - \delta(\omega' - \omega)] ,$$

(7)
it is clear that in this case Eq.(4) contains not only oscillating terms, but also a time-independent contribution,

\[ I_0^{(2)} = A_\omega [K^{(2)}(\omega, -\omega) + K^{(2)}(-\omega, \omega)] , \]  

where

\[ A_\omega = \frac{(-e)(2\pi \phi_\omega)^2}{4(Lm)^3 \phi_0^2} , \]  

and

\[ K^{(2)}(\omega, -\omega) = \sum_{\alpha \beta \gamma} \frac{P_{\alpha \beta \gamma}}{\varepsilon_\gamma - \varepsilon_\alpha + i0} \times \left[ \frac{f(\varepsilon_\gamma) - f(\varepsilon_\beta)}{\varepsilon_\gamma - \varepsilon_\beta - \omega + i0} - \frac{f(\varepsilon_\gamma) - f(\varepsilon_\alpha)}{\varepsilon_\beta - \varepsilon_\alpha + \omega + i0} \right] . \]  

Defining retarded and advanced Green’s functions,

\[ G^R_\alpha(\varepsilon) = \frac{1}{\varepsilon - \varepsilon_\alpha + i0} , \quad G^A_\alpha(\varepsilon) = \frac{1}{\varepsilon - \varepsilon_\alpha - i0} , \]  

Eq.(10) can also be written as

\[ K^{(2)}(\omega, -\omega) = -\frac{1}{2\pi i} \sum_{\alpha \beta \gamma} P_{\alpha \beta \gamma} \times \left\{ \int_\infty^{\infty} d\varepsilon \left[ f(\varepsilon + \omega) \left[ G^R_\alpha(\varepsilon + \omega) G^R_\beta(\varepsilon) G^R_\gamma(\varepsilon + \omega) \right] - G^A_\alpha(\varepsilon + \omega) G^A_\beta(\varepsilon) G^A_\gamma(\varepsilon + \omega) \right] \ight. 
\[ - \int_\infty^{\infty} d\varepsilon \left[ f(\varepsilon + \omega) - f(\varepsilon) \right] \times \left[ G^R_\alpha(\varepsilon + \omega) G^A_\beta(\varepsilon) G^A_\gamma(\varepsilon + \omega) \right] \left. - G^R_\alpha(\varepsilon + \omega) G^R_\beta(\varepsilon) G^A_\gamma(\varepsilon + \omega) \right) \right\} . \]  

The structure of the Green’s functions agrees with the one given by KY in Ref. [13]. Note, however, that these authors work in a different gauge: they represent the electric field by a scalar potential, so that their expressions contain only a single current vertex. The introduction of Green’s function is useful for calculating disorder averages. It is common wisdom that for the calculation of the disorder average of Eq.(12) the terms involving products of only retarded or only advanced Green’s functions can be neglected [12]. In this approximation a perturbative calculation of the disorder average of Eq.(12) has been given by KY [3], with the result that the associated time-independent part of the non-equilibrium current is proportional to \( \omega^{-2} \) for frequencies larger than the Thouless energy. As explained in Sec.3, for frequencies \( \omega < E_c \) the perturbative expansion is not controlled anymore since the energy spectrum becomes discrete. In fact, it will turn out, that the physical behavior is completely different in this regime.
To demonstrate the break down of the diagrammatic perturbation theory for systems with a discrete spectrum, we now show that an exact evaluation of the disorder average of Eq.(12) should actually yield an infinite result. Let us therefore go back to the exact spectral representation (10) of the response function. Using the formal identity

$$\frac{1}{x + i\delta} = \varphi \frac{1}{x} - i\pi\delta(x),$$

where \(\varphi\) denotes the Cauchy principal part, we can rewrite Eq.(10) as

$$K^{(2)}(\omega, -\omega) = K_{\varphi}^{(2)}(\omega, -\omega) + K_{\delta\delta}^{(2)}(\omega, -\omega),$$

with

$$K_{\varphi}^{(2)}(\omega, -\omega) = 2 \sum_{\alpha\beta\gamma} \text{Re} P_{\alpha\beta\gamma} f(\varepsilon_\gamma) - f(\varepsilon_\beta) \varepsilon_\gamma - \varepsilon_\beta - \omega,$$

$$K_{\delta\delta}^{(2)}(\omega, -\omega) = -2\pi^2 \sum_{\alpha\beta\gamma} \text{Re} P_{\alpha\beta\gamma} [f(\varepsilon_\gamma) - f(\varepsilon_\beta)] \times \delta(\varepsilon_\gamma - \varepsilon_\alpha) \delta(\varepsilon_\gamma - \varepsilon_\beta - \omega).$$

The terms with \(\alpha = \gamma\) in Eqs. (13) and (14) yield the following contributions,

$$K_{\varphi,\text{diag}}^{(2)}(\omega, -\omega) = \varphi \sum_{\alpha\beta} P_{\alpha\beta\alpha} \frac{\partial}{\partial \varepsilon_\alpha} \frac{f(\varepsilon_\alpha) - f(\varepsilon_\beta)}{\varepsilon_\alpha - \varepsilon_\beta - \omega},$$

$$= \varphi \sum_{\alpha\beta} P_{\alpha\beta\alpha} \left[ \frac{\partial}{\partial \varepsilon_\alpha} \frac{f(\varepsilon_\alpha) - f(\varepsilon_\beta)}{\varepsilon_\alpha - \varepsilon_\beta - \omega} \right],$$

$$K_{\delta\delta,\text{diag}}^{(2)}(\omega, -\omega) = -2\pi^2 \delta(0) \sum_{\alpha\beta} \text{Re} P_{\alpha\beta\alpha} [f(\varepsilon_\alpha) - f(\varepsilon_\beta)] \times \delta(\varepsilon_\alpha - \varepsilon_\beta - \omega).$$

The right-hand side of Eq.(18) is proportional to the infinite factor \(\delta(0)\). Hence, the term \(K_{\delta\delta}^{(2)}(\omega, -\omega)\) must also be infinite. Because the singular prefactor \(\delta(0)\) in Eq.(18) does not depend on the disorder, this singularity survives disorder averaging [14]. Keeping in mind that Eq.(12) is mathematically equivalent with Eq.(10), we conclude that a correct evaluation of the disorder average \(K^{(2)}(\omega, -\omega)\) must yield an infinite result [14]. Unfortunately, in an approximate evaluation of Eq.(12) by means of the usual diagrammatic methods this \(\delta\)-function singularity is artificially smoothed out, and one obtains a finite result [5].

### III. ADIABATIC SWITCHING ON

The infinite term (18) is clearly unphysical. This term is closely related to the infinitesimal imaginary parts \(i\delta\) that have been added to the real frequencies in the spectral representation (10) for the response function \(K^{(2)}(\omega, -\omega)\). As emphasized by KY [13], the
infinitesimal imaginary parts are a consequence of the fact that the response function must be causal when the time-dependent part of the Hamiltonian is adiabatically switched on. Let us examine the "adiabatic switching on" of the time-dependent perturbation more carefully. Following the usual recipe [15], we replace the Hamiltonian $\hat{H}_0 + \hat{V}(t)$ by $\hat{H}_0 + \hat{V}_\eta(t)$, where $\hat{V}_\eta(t) = \exp(\eta t)\hat{V}(t)$. The limit $\eta \to 0$ is then taken at the end of the calculation of physical quantities. For large enough times $t$ the physical result should be independent of the switching on procedure. Indeed, in the appendix we show by explicit calculation that sudden switching on produces the same result for the long-time response as adiabatic switching on. However, in the latter case one still has to be careful to take the limit $\eta \to 0$ only after the physical quantity of interest has been calculated. We now show that the singularity in Eq.(18) has been artificially generated by taking the limit $\eta \to 0$ at an intermediate step of the calculation.

By direct expansion of the time evolution operator in the interaction representation to second order in the time-dependent perturbation, we obtain the current for adiabatic switching on with finite $\eta$

$$I^{(2)}_\eta(t) = \frac{(-e)(2\pi)^2}{(mL)^3} \int_\infty^\infty d\omega_1 d\omega_2 \delta \varphi_1 \delta \varphi_2 \times K^{(2)}_{\eta t}(\omega_1, \omega_2)e^{-i(\omega_1+\omega_2)t},$$

with

$$K^{(2)}_{\eta t}(\omega_1, \omega_2) = e^{2\eta t} \sum_{\alpha\beta\gamma} \frac{P_{\alpha\beta\gamma}}{\varepsilon_\gamma - \varepsilon_\alpha + \omega_1 + \omega_2 + 2i\eta}$$

$$\times \left[ \frac{f(\varepsilon_\gamma) - f(\varepsilon_\beta)}{\varepsilon_\gamma - \varepsilon_\beta + \omega_2 + i\eta} - \frac{f(\varepsilon_\beta) - f(\varepsilon_\alpha)}{\varepsilon_\beta - \varepsilon_\alpha + \omega_1 + i\eta} \right].$$

Comparing Eq.(21) with Eq.(5), we see that the former is multiplied by an extra factor of $e^{2\eta t}$. If we directly take the limit $\eta \to 0$, this factor is replaced by unity. This is the limiting procedure adopted in the usual Green’s function approach, where one takes first the limit $\eta \to 0$ in Eq.(21) and then inserts the resulting expression into Eq.(19). In this case we recover Eqs.(8) and (14), which lead to the divergence in Eq.(18). We now show that this unphysical divergence does not appear if the limit $\eta \to 0$ is taken after the physical current has been calculated. Substituting Eq.(21) into Eq.(19) we obtain

$$I^{(2)}_\eta(t) = A_\omega [K^{(2)}_{\eta t}(\omega, -\omega) + K^{(2)}_{\eta t}(-\omega, \omega)$$

$$+ K^{(2)}_{\eta t}(\omega, \omega)e^{-2i\omega t} + K^{(2)}_{\eta t}(-\omega, -\omega)e^{2i\omega t}].$$

In analogy with Eq.(14), we express $K^{(2)}_{\eta t}(\omega, -\omega)$ in terms of products of real and imaginary parts

$$K^{(2)}_{\eta t}(\omega, -\omega) = K^{(2)}_{\eta t,\Re}(\omega, -\omega) + K^{(2)}_{\eta t,\Im}(\omega, -\omega),$$

with
\[ K^{(2)}_{\eta,\varphi}(\omega, -\omega) = 2 e^{2\varphi t} \sum_{\alpha\beta\gamma} \text{Re} P_{\alpha\beta\gamma} [f(\varepsilon_{\gamma}) - f(\varepsilon_{\beta})] \]
\[ \times \left[ \frac{\varepsilon_{\gamma} - \varepsilon_{\alpha}}{(\varepsilon_{\gamma} - \varepsilon_{\alpha})^2 + (2\eta)^2} \frac{\varepsilon_{\gamma} - \varepsilon_{\beta} - \omega}{(\varepsilon_{\gamma} - \varepsilon_{\beta} - \omega)^2 + \eta^2} \right] , \] (23)

\[ K^{(2)}_{\eta,\delta\delta}(\omega, -\omega) = -2 e^{2\varphi t} \sum_{\alpha\beta\gamma} \text{Re} P_{\alpha\beta\gamma} [f(\varepsilon_{\gamma}) - f(\varepsilon_{\beta})] \]
\[ \times \left[ \frac{2\eta}{(\varepsilon_{\gamma} - \varepsilon_{\alpha})^2 + (2\eta)^2} \frac{\eta}{(\varepsilon_{\gamma} - \varepsilon_{\beta} - \omega)^2 + \eta^2} \right] . \] (24)

From Eq. (23) it is now obvious that \( K^{(2)}_{\eta,\varphi} \) does not have any contributions from the terms \( \alpha = \gamma \). The finite contribution in Eq. (17) is thus an artifact of taking the limit \( \eta \to 0 \) before calculating any physical quantities. Let us now focus on the term (24). If we directly take the limit \( \eta \to 0 \) using
\[ \lim_{\eta \to 0} \frac{\eta}{\varepsilon^2 + \eta^2} = \pi \delta(\epsilon) , \] (25)
we recover the infinite result (18). However, the structure of the \( \eta \)-dependent part of Eq. (24) is familiar from the derivation of Fermi’s golden rule of elementary quantum mechanics. As discussed for example in the classic textbook by Baym [15], terms with this structure should be interpreted as a rate, i.e. as a contribution to the current that grows linearly in time. It is therefore clear that after taking the derivative of Eq. (24) with respect to \( t \) we obtain a finite result if we then let \( \eta \to 0 \). A simple calculation yields
\[ \lim_{\eta \to 0} \frac{d}{dt} K^{(2)}_{\eta,\delta\delta}(\omega, -\omega) \]
\[ = -2 \lim_{\eta \to 0} \sum_{\alpha\beta} P_{\alpha\beta\alpha} \frac{[f(\varepsilon_{\alpha}) - f(\varepsilon_{\beta})] \eta}{(\varepsilon_{\alpha} - \varepsilon_{\beta} - \omega)^2 + \eta^2} \]
\[ = -2\pi \sum_{\alpha\beta} P_{\alpha\beta\alpha} [f(\varepsilon_{\alpha}) - f(\varepsilon_{\beta})] \delta(\varepsilon_{\alpha} - \varepsilon_{\beta} - \omega) . \] (26)

Because this expression contains only a single \( \delta \)-function, after averaging over disorder it becomes a smooth function of \( \omega \). We conclude that to quadratic order in the field the non-equilibrium current induced by the time-dependent flux (1) has the following three contributions,
\[ I^{(2)}(t) \equiv \lim_{\eta \to 0} I^{(2)}_{\eta}(t) = I^{(2)}_{\text{th}} + t \frac{dI^{(2)}_{\text{kin}}}{dt} + I^{(2)}_{\text{osc}}(t) , \] (27)

where the time-independent part is given by
\[ I^{(2)}_{\text{th}} = A_\omega \lim_{\eta \to 0} [K^{(2)}_{\eta,\delta\delta}(\omega, -\omega) + K^{(2)}_{\eta,\varphi}(-\omega, \omega)] \]
\[ = 2A_\omega \sum_{\alpha\gamma, \alpha \neq \gamma} \text{Re} P_{\alpha\beta\gamma} \]
\[ \times \varphi \left[ \frac{f(\varepsilon_{\gamma}) - f(\varepsilon_{\beta})}{\varepsilon_{\gamma} - \varepsilon_{\beta} - \omega} + (\omega \to -\omega) \right] . \] (28)
The coefficient of the term linear in time is
\[
\frac{dI^{(2)}_{\text{kin}}}{dt} = A_\omega \lim_{\eta \to 0} \left[ \frac{d}{dt} K^{(2)}_{\eta t, \delta \delta} (\omega, -\omega) + \frac{d}{dt} K^{(2)}_{\eta t, \delta \delta} (-\omega, \omega) \right]
\]
\[
= -2\pi A_\omega \sum_{\alpha \beta} P_{\alpha \beta \alpha} [f(\varepsilon_\alpha) - f(\varepsilon_\beta)]
\times \left[ \delta(\varepsilon_\alpha - \varepsilon_\beta - \omega) + (\omega \to -\omega) \right],
\]  
(29)
and the oscillating part is
\[
I^{(2)}_{\text{osc}}(t) = A_\omega \lim_{\eta \to 0} [K^{(2)}_{\eta t}(\omega, \omega)e^{-2i\omega t} + K^{(2)}_{\eta t}(-\omega, -\omega)e^{2i\omega t}].
\]  
(30)

Thus, a time-dependent electric field with frequency \(\omega\) induces in quadratic order three fundamentally different currents. (a) A time-independent contribution \(I^{(2)}_{\text{th}}\); as shown in the next section, this contribution can be derived from a thermodynamic calculation. (b) A contribution \(tdI^{(2)}_{\text{kin}}/dt\) which increases linearly in time; this term can be understood in terms of the usual golden rule of time-dependent perturbation theory. (c) Finally, there is also a time-dependent contribution \(I^{(2)}_{\text{osc}}\) oscillating with frequency \(2\omega\). When this term is averaged over a time-interval larger than \(\omega^{-1}\), its contribution to the current is negligibly small.

From the above analysis it is clear that the contribution that is proportional to \(t\) cannot be calculated within the usual Green’s function machinery, because in this approach the limit \(\eta \to 0\) is taken at an intermediate step of the calculation, causing an unphysical divergence. To further support the correctness of the limiting procedure adopted here we show in the appendix that Eqs. (27–29) can also be re-derived if the perturbation is suddenly (instead of adiabatically) switched on.

**IV. THE THERMODYNAMIC ORIGIN OF THE TIME-INDEPENDENT PART OF THE CURRENT**

The time-independent part \(I^{(2)}_{\text{th}}\) of the non-equilibrium current in Eq. (27) has been discussed by us in Ref. [16]. This contribution can be obtained from a thermodynamic calculation. In Ref. [16] we have assumed (without further justification) the existence of such a relation. Let us now put this assumption on a more solid basis. Within the Matsubara (imaginary time) formalism one can directly calculate the imaginary frequency version of the response function \(K^{(2)}(\omega_1, \omega_2)\) given in Eq. (5), i.e.
\[
K^{(2)}(\omega_1, \omega_2) = \sum_{\alpha \beta \gamma} \frac{P_{\alpha \beta \gamma}}{\varepsilon_\gamma - \varepsilon_\alpha + i\omega_1 + i\omega_2}
\times \left[ \frac{f(\varepsilon_\gamma) - f(\varepsilon_\beta)}{\varepsilon_\gamma - \varepsilon_\beta + i\omega_2} - \frac{f(\varepsilon_\beta) - f(\varepsilon_\alpha)}{\varepsilon_\beta - \varepsilon_\alpha + i\omega_1} \right].
\]  
(31)

As pointed out by KY [13], in order to obtain the causal response function, one should first continue both frequencies to the real axis with positive imaginary part \((i\omega_1 \to \omega_1 + i0, i\omega_2 \to \omega_2 + i0)\), and then set \(\omega_1 = -\omega_2\) to obtain the constant part of the physical current.
On the other hand, if one performs these steps in opposite order (i.e. first sets $i\omega_1 = -i\omega_2$ and then continues $i\omega_1 \rightarrow \omega + i0$) one obtains for the current response function

$$K_{\text{th}}^{(2)}(\omega, -\omega) = \text{Re} \sum_{\alpha\beta\gamma} P_{\alpha\beta\gamma} \frac{\epsilon_\gamma - \epsilon_\alpha}{\epsilon_\gamma - \epsilon_\beta - \omega - i0} \left[ f(\epsilon_\gamma) - f(\epsilon_\beta) - f(\epsilon_\alpha) \right].$$

Comparing this expression with Eqs.(14–16), it is easy to see that

$$K_{\text{th}}^{(2)}(\omega, -\omega) = K_{\text{th}}^{(2)}(\omega, -\omega).$$

Hence, the time-independent part $I_{\text{th}}^{(2)}$ of the current can indeed be obtained from a thermodynamic calculation [16]. Note, however, that our analysis of Sec.III (see also the appendix) implies that the terms with $\alpha = \gamma$ in Eq.(32) should be omitted from the sum, i.e. the physical current is given by

$$I_{\text{th}}^{(2)} = A_\omega \left[ \tilde{K}_{\text{th}}^{(2)}(\omega, -\omega) + \tilde{K}_{\text{th}}^{(2)}(-\omega, \omega) \right].$$

where

$$\tilde{K}_{\text{th}}^{(2)}(\omega, -\omega) = K_{\text{th}}^{(2)}(\omega, -\omega) - K_{\text{th}}^{(2)}(\omega, \omega)$$

$$= \sum_{\alpha\beta\gamma, \alpha \neq \gamma} P_{\alpha\beta\gamma} \frac{\epsilon_\gamma - \epsilon_\alpha}{\epsilon_\gamma - \epsilon_\beta - \omega} \left[ f(\epsilon_\gamma) - f(\epsilon_\beta) - f(\epsilon_\alpha) \right].$$

(35)

see Eq.(17). The direct diagrammatic calculation of the disorder average of $I_{\text{th}}^{(2)}$ is difficult, because the restriction $\alpha \neq \gamma$ in Eq.(33) is not so easy to implement. In Ref. [16] the following limiting procedure was adopted: Instead of directly calculating $K_{\text{th}}^{(2)}(\omega, -\omega)$, consider the generalization of the imaginary frequency response function (31) for electric fields with finite wave-vector $q$, which we denote by $K^{(2)}(i\omega, -i\omega, q)$. The limit $q \rightarrow 0$ is taken after the disorder averaged current has been calculated. As shown in Ref. [16], in the diffusive regime the function $K^{(2)}(i\omega, -i\omega, q)$ is a smooth function of $q$, so that the limit $q \rightarrow 0$ is well defined. The so-defined averaged response function vanishes for frequencies exceeding the Thouless energy as $\exp(-\sqrt{|\omega|/2E_c})$ [16]. On the other hand, perturbative averaging of the contribution from the (unwanted) diagonal term (17) shows that this term vanishes as $\omega^{-2}$ for large frequencies. This indicates that the above limiting procedure indeed eliminates the contribution of the unphysical diagonal term (17) to the time-independent part of the non-equilibrium current.

V. CONCLUSION

In this work we have shown that a time-dependent flux oscillating with frequency $\omega$ that pierces the center of a mesoscopic metal ring generates to quadratic order three fundamentally different contributions to the current: a constant non-equilibrium current $I_{\text{th}}^{(2)}$,
a current $tdI_{\text{kin}}^{(2)}/dt$ that grows linearly in time, and a current oscillating with frequency $2\omega$.

As shown in Ref. [16], the disorder average of the constant term $I_{\text{th}}^{(2)}$ vanishes for frequencies exceeding the Thouless energy as $\exp(-\sqrt{|\omega|/2E_c})$. The calculation of the disorder average of the contribution $tdI_{\text{kin}}^{(2)}/dt$ remains an open problem. A direct perturbative calculation by means of the impurity diagram technique is not straightforward, because Eq.(29) involves three matrix elements but only one energy denominator. Therefore this expression cannot be simply written in terms of Green’s functions.

The main result of this work is the prediction of a current $tdI_{\text{kin}}^{(2)}/dt$ increasing linearly with time. From the well-known derivation of Fermi’s golden rule [15] it is clear that this result is only valid in an intermediate time interval. In particular, the calculation of the long-time behavior of the non-equilibrium current requires non-perturbative methods.

One should keep in mind that our calculation has been performed for non-interacting electrons in a random potential, so that our results are valid as long as the spectrum of the system is discrete. We have argued in Sec. I that at low enough temperatures this should be the case for small external frequencies, $|\omega| < E_c$. On the other hand, for frequencies exceeding $E_c$ the spectrum is effectively continuous. In this regime the conventional Green’s function methods can be used to calculate the direct current, so that the results of KY [3] should be valid.

Let us also point out that the linear time-dependence of the current is a consequence of the discrete spectrum, and is not related to the adiabatic switching on procedure in Eq.(20). In the appendix we show that sudden switching on yields the same linear time-dependence of the current. It seems reasonable to expect that for sufficiently short times the constant part $I_{\text{th}}^{(2)}$ of the current is dominant [14]. We would like to encourage experimentalists to measure the non-equilibrium response of mesoscopic metal rings to a time-dependent flux in the frequency range $10^8\text{Hz} \leq \omega \leq 10^{13}\text{Hz}$.

ACKNOWLEDGEMENT

This work was supported by the Deutsche Forschungsgemeinschaft (SFB 345). We thank V. E. Kravtsov for his comments.

APPENDIX: SUDDEN SWITCHING ON

To confirm that the ”switching on procedure” outlined in Sec.III yields the correct physical results, let us consider a harmonic perturbation that is suddenly turned on at time $t = 0$,

$$\phi(t) = \phi + \phi_\omega \Theta(t) \sin(\omega t),$$  \hspace{1cm} (A1)

where $\Theta(t)$ is the step function. To second order in $\phi_\omega$ the induced current is

$$I^{(2)}(t) = 2A_{\omega} \text{Re} \sum_{\alpha\beta\gamma} P_{\alpha\beta\gamma} [f(\varepsilon_\beta) - f(\varepsilon_\alpha)]$$
\[
\times \left[ \frac{e^{2i\omega t} - e^{i(\varepsilon_{\gamma} - \varepsilon_{\alpha})t}}{(\varepsilon_{\alpha} - \varepsilon_{\gamma} + 2\omega)(\varepsilon_{\alpha} - \varepsilon_{\beta} + \omega)} \right. \\
\frac{1 - e^{i(\varepsilon_{\alpha} - \varepsilon_{\gamma})t}}{(\varepsilon_{\alpha} - \varepsilon_{\gamma})(\varepsilon_{\alpha} - \varepsilon_{\beta} - \omega)} \\
+ \frac{2\omega}{(\varepsilon_{\alpha} - \varepsilon_{\beta})^2 - \omega^2} \left[ \frac{e^{i(\varepsilon_{\beta} - \varepsilon_{\alpha} + \omega)t} - e^{i(\varepsilon_{\gamma} - \varepsilon_{\alpha})t}}{\varepsilon_{\beta} - \varepsilon_{\gamma} + \omega} \right] \\
\left. + (\omega \rightarrow -\omega) \right].
\] (A2)

The diagonal term \(\alpha = \gamma\) is

\[
I^{(2)}_{\text{diag}}(t) = 4A_\omega \sum_{\alpha\beta} P_{\alpha\beta\alpha} [f(\varepsilon_{\alpha}) - f(\varepsilon_{\beta})] \\
\times \left[ \frac{\sin^2(\omega t)}{(\varepsilon_{\alpha} - \varepsilon_{\beta})^2 - \omega^2} \\
- \frac{\sin^2(\varepsilon_{\beta} - \varepsilon_{\alpha} + \omega t) + \sin^2(\frac{\varepsilon_{\beta} - \varepsilon_{\alpha} - \omega t}{2})}{(\varepsilon_{\alpha} - \varepsilon_{\beta})^2 - \omega^2} \\
+ \left[ \frac{\sin(\varepsilon_{\beta} - \varepsilon_{\alpha} + \omega t)}{\varepsilon_{\beta} - \varepsilon_{\alpha} + \omega} \right]^2 + \left[ \frac{\sin(\frac{\varepsilon_{\beta} - \varepsilon_{\alpha} - \omega t}{2})}{\varepsilon_{\beta} - \varepsilon_{\alpha} - \omega} \right]^2 \right].
\] (A3)

The terms in the last line can be interpreted in the same way as is done in Fermi’s golden rule [15] by using the identity

\[
\left[ \frac{\sin \left( \frac{\Delta \varepsilon t}{2} \right)}{\Delta \varepsilon} \right]^2 \rightarrow \frac{\pi}{2} t \delta(\Delta \varepsilon) \text{ for } t \rightarrow \infty.
\] (A4)

It is now easy to see that for large times \(I^{(2)}_{\text{diag}}(t)\) yields exactly the same linear in time contribution as given in Eq.(29). The terms with no explicit time dependence in Eq.(A2) can be identified with \(I^{(2)}_{\text{th}}\) in Eq.(28).
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