

Non-linear response and electron-electron interactions in mesoscopic metal rings

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Abstract. A time-dependent electric field gives rise to a stationary non-equilibrium current $I^{(2)}$ around a mesoscopic metal ring threaded by a magnetic flux. We show that this current, which is proportional to the intensity of the field, is closely related to the exchange part of the interaction contribution to the *equilibrium* persistent current, and that the corresponding non-linear conductivity directly measures the weak localization correction to the polarization. We explicitly calculate the disorder average of $I^{(2)}$ in the diffusive regime as function of the frequency of the electric field and the static flux piercing the ring, and suggest an experiment to test our theory.

PACS. 73.50.Bk General theory, scattering mechanisms – 72.10.Bg General formulation of transport theory – 72.15.Rn Quantum localization

1 Introduction

Electron-electron interactions in disordered mesoscopic metals are not very well understood. The usual perturbative machinery of many-body theory is not always applicable in these systems, because often the intricate interplay between interactions and disorder in phase coherent systems cannot be described by means of simple perturbation theory. The persistent current [1] in a mesoscopic diffusive metal ring threaded by a magnetic flux belongs to this category. Seven years after the seminal experiment by Lévy *et al.* [2], there seems to be general agreement that electron-electron interactions are essential to explain the surprisingly large magnitude of the experimentally measured average persistent current in an array of 10^7 Curing. Note that the experiment by Lévy *et al.* has recently been independently confirmed [3].

In this work we shall study electron-electron interactions in mesoscopic metal rings by means of a somewhat unconventional approach, which is based on the connection between electron-electron interactions on the one hand, and non-linear response to an external electromagnetic field on the other hand. In the context of persistent currents this connection has recently been pointed out by Kravtsov and Yudson [4], who considered the *time-independent* part of the non-equilibrium current proportional to the intensity of an external longitudinal electric field $E(t) = \Re[E(\omega)e^{i\omega t}]$,

$$I^{(2)} = \Re \left[\sigma^{(2)}(\omega, \phi) \right] |E(\omega)|^2. \quad (1)$$

The so defined non-linear conductivity $\sigma^{(2)}(\omega, \phi)$ (see Eqs. (47, 48) below) is a function of the frequency ω of

the external electric field, as well as of the static flux ϕ piercing the ring. The fact that non-linear response and interactions are closely related becomes obvious in a path-integral approach. Indeed, it is well-known [5] that the Coulomb interaction between electrons can be obtained by integrating the exponential of the coupled Maxwell-matter action over the fluctuating quantum electric and magnetic fields. However, in a path integral approach we may also perform the integrations in a different order. Thus, an alternative method to obtain the equilibrium current is to calculate first the non-equilibrium current for a given realization of the electromagnetic fields, and then performing an average over these fields. The effective action for this averaging procedure is obtained by integrating first over the electronic degrees of freedom, keeping the electromagnetic fields fixed. In this work we shall show that this procedure leads to new perspectives in the role of electron-electron interactions for persistent currents, which can be tested experimentally by means of non-linear transport experiments. We shall also re-examine an earlier calculation of the non-equilibrium current (1) due to Kravtsov and Yudson [4].

The equivalence between Coulomb interactions and fluctuating electromagnetic fields has been employed previously by Altshuler, Aronov and Khmelnitsky [6] in their calculation of the dephasing rate due to electron-electron interactions in disordered metals. See also reference [7] for a recent study of interaction effects in mesoscopic conductors with the help of this approach.

The plan of the rest of this paper is as follows: in Section 2 we shall use well-known functional techniques to show how the above averaging procedure can be carried out in practice, and elucidate the precise connection

between non-linear response and electron-electron interactions. In particular, we show that the non-equilibrium current (1) is closely related to the Fock contribution to the equilibrium persistent current, and that the non-linear conductivity $\sigma^{(2)}(\omega, \phi)$ in equation (1) can be simply obtained from the weak localization correction to the polarization of the system. In Section 3 we shall explicitly evaluate the average non-equilibrium current $\bar{I}^{(2)}$ as function of ω and ϕ , and compare our result with reference [4] (here and below the over-bar denotes averaging over the disorder). In Section 4 we discuss possibilities to test our theory experimentally, and conclude in Section 5 with a brief summary.

2 From interactions to non-linear response

In this section we shall map the problem of calculating the equilibrium persistent current of interacting electrons onto an effective non-equilibrium problem in imaginary time. This is achieved by means of a Hubbard-Stratonovich transformation, a well known technique in the theory of strongly correlated electrons [8,9].

2.1 Definition of the problem

We consider a system consisting of electrons with charge $-e$ and mass m which are confined to a thin ring with circumference L and cross section L_{\perp}^2 , with $L_{\perp} \ll L$. The electrons interact with two-body Coulomb forces and move in a static random potential $U(\mathbf{r})$. Identifying the position along the circumference of the ring with the x -coordinate, the equilibrium current I around the ring can be written as

$$I = \frac{-e}{L} \int d\mathbf{r} j(\mathbf{r}), \quad (2)$$

where the integral is over the volume $\mathcal{V} = LL_{\perp}^2$ of the ring, and the particle current density $j(\mathbf{r})$ (in x -direction) can be expressed in terms of the exact imaginary time Green's function $G(\mathbf{r}, \mathbf{r}', \tau - \tau')$ as

$$\begin{aligned} j(\mathbf{r}) &= \lim_{\tau' \rightarrow \tau} \lim_{\mathbf{r}' \rightarrow \mathbf{r}} \hat{J}_{x,x'} G(\mathbf{r}, \mathbf{r}', \tau - \tau' - 0^+) \\ &= T \sum_{n=-\infty}^{\infty} e^{i\tilde{\omega}_n 0^+} \lim_{\mathbf{r}' \rightarrow \mathbf{r}} \hat{J}_{x,x'} G(\mathbf{r}, \mathbf{r}', i\tilde{\omega}_n), \end{aligned} \quad (3)$$

with the differential operator

$$\hat{J}_{x,x'} \equiv \frac{1}{2mi} (\partial_x - \partial_{x'}) + \frac{a}{m}, \quad a = \frac{2\pi}{L} \frac{\phi}{\phi_0}. \quad (4)$$

Here $\phi_0 = 2\pi c/e$ is the flux quantum, and we have introduced the imaginary frequency Fourier transform of the Green's function,

$$G(\mathbf{r}, \mathbf{r}', \tau - \tau') = T \sum_{n=-\infty}^{\infty} e^{-i\tilde{\omega}_n(\tau - \tau')} G(\mathbf{r}, \mathbf{r}', i\tilde{\omega}_n), \quad (5)$$

where $\tilde{\omega}_n = 2\pi(n + \frac{1}{2})T$. For simplicity, we shall work with spinless electrons and use units where \hbar and the Boltzmann-constant are set equal to unity. This amounts to measuring temperatures T and frequencies ω in units of energy. The Green's function at constant chemical potential μ can be represented as a functional integral over Grassmann fields ψ and ψ^\dagger in the usual way [9],

$$G(\mathbf{r}, \mathbf{r}', \tau - \tau') = - \frac{\int \mathcal{D}\{\psi\} e^{-S\{\psi\}} \psi(\mathbf{r}, \tau) \psi^\dagger(\mathbf{r}', \tau')}{\int \mathcal{D}\{\psi\} e^{-S\{\psi\}}}, \quad (6)$$

where $S\{\psi\} = S_0\{\psi\} + S_{int}\{\psi\}$, with

$$S_0\{\psi\} = -\frac{1}{T} \sum_{kk'} \psi_k^\dagger [\hat{G}_0^{-1}]_{kk'} \psi_{k'}, \quad (7)$$

$$S_{int}\{\psi\} = \frac{1}{2T} \sum_q f_{\mathbf{q}} \rho_{-q} \rho_q. \quad (8)$$

Here $\rho_q = \sum_k \psi_k^\dagger \psi_{k+q}$, and the inverse non-interacting Green's function matrix in the momentum-frequency basis for a given realization of the disorder potential is

$$[\hat{G}_0^{-1}]_{kk'} = \delta_{kk'} \left[i\tilde{\omega}_n - \frac{(\mathbf{k} + \mathbf{a})^2}{2m} + \mu \right] - \delta_{nn'} U_{\mathbf{k}-\mathbf{k}'}, \quad (9)$$

where \mathbf{a} is a vector potential directed along the circumference of the ring (which in our convention is identified with the x -direction), with magnitude $a \equiv |\mathbf{a}| = \frac{2\pi}{L} \frac{\phi}{\phi_0}$. For simplicity we have introduced collective labels $k = [\mathbf{k}, i\tilde{\omega}_n]$ and $q = [\mathbf{q}, i\omega_n]$ for wave-vector and Matsubara frequencies, where $\tilde{\omega}_n = 2\pi(n + \frac{1}{2})T$ is a fermionic frequency, and $\omega_n = 2\pi nT$ is a bosonic one. The Grassmann variables ψ_k are the Fourier components of the field $\psi(\mathbf{r}, \tau)$, *i.e.* $\psi(\mathbf{r}, \tau) = \mathcal{V}^{-1/2} \sum_k e^{i(\mathbf{k}\cdot\mathbf{r} - \tilde{\omega}_n\tau)} \psi_k$. The Fourier transforms $U_{\mathbf{q}}$ of the disorder potential and $f_{\mathbf{q}}$ of the Coulomb potential are normalized such that both have units of energy [10], *i.e.*

$$U_{\mathbf{q}} = \frac{1}{\mathcal{V}} \int d\mathbf{r} e^{-i\mathbf{q}\cdot\mathbf{r}} U(\mathbf{r}), \quad (10)$$

$$f_{\mathbf{q}} = \frac{1}{\mathcal{V}^2} \int d\mathbf{r} d\mathbf{r}' e^{-i\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}')} \frac{e^2}{|\mathbf{r}-\mathbf{r}'|}. \quad (11)$$

The disorder potential $U(\mathbf{r})$ is assumed to have zero average and Gaussian white noise correlations, so that

$$\overline{U_{\mathbf{q}} U_{\mathbf{q}'}} = \tilde{\gamma} \delta_{\mathbf{q}, -\mathbf{q}'}, \quad (12)$$

where the parameter $\tilde{\gamma}$ is a measure for the strength of the disorder. Within lowest order Born approximation we may identify $\tilde{\gamma} = \Delta/(2\pi\tau)$, where Δ is the average level spacing at the Fermi energy, and τ is the elastic lifetime.

The evaluation of the above expression for the current would require the solution of the many-body problem in the presence of disorder, an impossible task. Perturbative expansions can be performed in powers of the disorder potential $U_{\mathbf{q}}$ and in powers of the Coulomb interaction $f_{\mathbf{q}}$. This double expansion is rather subtle. To obtain sensible

results which correctly take into account the physics of diffusion and screening, infinitely many powers of $U_{\mathbf{q}}$ and $f_{\mathbf{q}}$ have to be summed. In order to make this expansion more transparent and to see the connection with non-linear response, we shall now map this problem onto an equivalent problem where the two-body interaction is replaced by a time-dependent auxiliary field.

2.2 Hubbard-Stratonovich transformation and equivalent non-equilibrium problem

The two-body part $S_{int}\{\psi\}$ of our effective action can be decoupled by means of the following Hubbard-Stratonovich transformation [8]

$$e^{-S_{int}\{\psi\}} = \frac{\int \mathcal{D}\{\Phi\} \exp \left[-\frac{T}{2} \sum_{\mathbf{q}} f_{\mathbf{q}}^{-1} \Phi_{-\mathbf{q}} \Phi_{\mathbf{q}} - i \sum_{\mathbf{q}} \Phi_{-\mathbf{q}} \rho_{\mathbf{q}} \right]}{\int \mathcal{D}\{\Phi\} \exp \left[-\frac{T}{2} \sum_{\mathbf{q}} f_{\mathbf{q}}^{-1} \Phi_{-\mathbf{q}} \Phi_{\mathbf{q}} \right]}. \quad (13)$$

Applying this transformation to the denominator and numerator of equation (6), and integrating over the Grassmann field, the exact current density of the many-body system can be written as

$$j(\mathbf{r}) = \frac{\int \mathcal{D}\{\Phi\} e^{-S_{eff}\{\Phi\}} j(\mathbf{r}, \tau, \{\Phi\})}{\int \mathcal{D}\{\Phi\} e^{-S_{eff}\{\Phi\}}} \equiv \langle j(\mathbf{r}, \tau, \{\Phi\}) \rangle_{S_{eff}}. \quad (14)$$

The effective action $S_{eff}\{\Phi\}$ is given by

$$S_{eff}\{\Phi\} = \frac{T}{2} \sum_{\mathbf{q}} f_{\mathbf{q}}^{-1} \Phi_{-\mathbf{q}} \Phi_{\mathbf{q}} - Tr \ln \left[1 - \hat{G}_0 \hat{V} \right], \quad (15)$$

where the matrix elements of the infinite matrix \hat{V} are given by $[\hat{V}]_{kk'} = V_{k-k'} = iT\Phi_{k-k'}$. The quantity $j(\mathbf{r}, \tau, \{\Phi\})$ is the non-equilibrium current density for a frozen configuration of the Hubbard-Stratonovich field, *i.e.*

$$j(\mathbf{r}, \tau, \{\Phi\}) = \lim_{\mathbf{r}' \rightarrow \mathbf{r}} \hat{J}_{x,x'} \mathcal{G}(\mathbf{r}, \mathbf{r}', \tau, \tau + 0^+). \quad (16)$$

Here \mathcal{G} satisfies the partial differential equation

$$\left[-\partial_{\tau} - \frac{(-i\nabla_{\mathbf{r}} + \mathbf{a})^2}{2m} + \mu - U(\mathbf{r}) - V(\mathbf{r}, \tau) \right] \mathcal{G}(\mathbf{r}, \mathbf{r}', \tau, \tau') = \delta(\mathbf{r} - \mathbf{r}') \delta^*(\tau - \tau'), \quad (17)$$

where the time-dependent potential $V(\mathbf{r}, \tau)$ is defined by

$$V(\mathbf{r}, \tau) = \sum_{\mathbf{q}} e^{i(\mathbf{q}\cdot\mathbf{r} - \omega_n \tau)} V_{\mathbf{q}}, \quad V_{\mathbf{q}} = iT\Phi_{\mathbf{q}}, \quad (18)$$

and $\delta^*(\tau) = T \sum_n e^{-i\tilde{\omega}_n \tau}$ is the antiperiodic imaginary time δ -function. Note that the potential $V(\mathbf{r}, \tau)$ is a periodic function of τ , *i.e.* $V(\mathbf{r}, \tau + 1/T) = V(\mathbf{r}, \tau)$. On the

other hand, the fermionic Green's function \mathcal{G} has to satisfy antiperiodic boundary conditions in each imaginary time variable [11],

$$\begin{aligned} \mathcal{G}(\mathbf{r}, \mathbf{r}', \tau + 1/T, \tau') &= -\mathcal{G}(\mathbf{r}, \mathbf{r}', \tau, \tau') \\ &= \mathcal{G}(\mathbf{r}, \mathbf{r}', \tau, \tau' + 1/T). \end{aligned} \quad (19)$$

The above transformation is exact, and allows us to clarify the precise connection between interactions and non-linear response [4]. Note that equation (17) defines the *imaginary time* non-equilibrium Green's function of non-interacting fermions subject to an external imaginary time potential $V(\mathbf{r}, \tau)$. Of course, for a comparison with experiments, which measure the non-linear response to external fields, we need to know the real time dynamics. While within linear response the well-known fluctuation-dissipation theorem tells us how to obtain the real time response by simple analytic continuation from an imaginary time formalism, in the case of non-linear response the situation is more complicated. Nevertheless, even then the analytic continuation from the imaginary time response to real times is possible, provided the time-dependence of the external potential can be analytically continued, and the potential is adiabatically switched on [12]. This point, which apparently is not widely appreciated in the literature, has already been discussed in the classic textbook by Kadanoff and Baym [11].

2.3 How functional averaging reproduces the equilibrium current

For a calculation of the equilibrium persistent current to first order in the RPA (random phase approximation) screened interaction, it is sufficient to expand the effective action (15) and the non-equilibrium current-density $j(\mathbf{r}, \tau, \{\Phi\})$ defined in equation (16) to second order in the Hubbard-Stratonovich field. The resulting Gaussian integrations can then be performed exactly. In this approximation the effective action is given by

$$\begin{aligned} S_{eff}\{\Phi\} &\approx i \sum_{\mathbf{q}} N_0(\mathbf{q}) \Phi_{-\mathbf{q}} \\ &+ \frac{T}{2} \sum_{\mathbf{q}\mathbf{q}'} [\delta_{\mathbf{q}\mathbf{q}'} f_{\mathbf{q}}^{-1} + \Pi_0(\mathbf{q}, \mathbf{q}')] \Phi_{-\mathbf{q}} \Phi_{\mathbf{q}'} + \dots, \end{aligned} \quad (20)$$

where

$$N_0(\mathbf{q}) = \delta_{n0} N_0(\mathbf{q}) = T \sum_k [\hat{G}_0]_{k+\mathbf{q}, k}, \quad (21)$$

$$\begin{aligned} \Pi_0(\mathbf{q}, \mathbf{q}') &= \delta_{nn'} \Pi_0(\mathbf{q}, \mathbf{q}', i\omega_n) \\ &= -T \sum_{kk'} [\hat{G}_0]_{k+\mathbf{q}, k'+\mathbf{q}'} [\hat{G}_0]_{k'k}. \end{aligned} \quad (22)$$

Physically $N_0(\mathbf{q})$ is the spatial Fourier component of the density, and $\Pi_0(\mathbf{q}, \mathbf{q}', i\omega_n)$ is the non-interacting polarization [10] for a given realization of the disorder potential $U(\mathbf{r})$. To expand the non-equilibrium current-density, it is

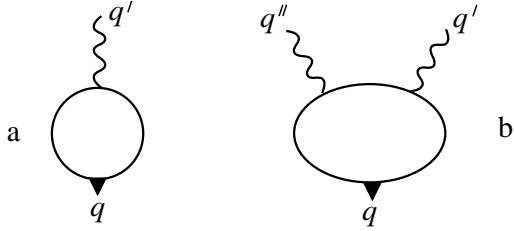


Fig. 1. (a) Graphical representation of the current-density $j_q^{(1)}$ in linear response, see equation (27). The solid lines represent non-interacting Green's functions for fixed disorder potential, the wavy lines represent the fields V_q , and the black triangle denotes the current vertex $(k_x + a + q_x/2)/m$. (b) Graphical representation of $j_q^{(2)}$.

convenient to consider the Fourier components,

$$j(\mathbf{r}, \tau, \{\Phi\}) = \frac{1}{V} \sum_{\mathbf{q}} e^{i(\mathbf{q}\cdot\mathbf{r} - \omega_n \tau)} j_{\mathbf{q}}. \quad (23)$$

Note that with this normalization the equilibrium current defined in equation (2) is simply given by

$$I = \frac{-e}{L} \langle j_{q=0} \rangle_{S_{eff}}, \quad (24)$$

where $q = 0$ means $\mathbf{q} = 0$ and $\omega_n = 0$. For j_q we obtain the following expansion in powers of $V_q = iT\Phi_q$,

$$j_q = j_q^{(0)} + j_q^{(1)} + j_q^{(2)} + \dots, \quad (25)$$

where

$$j_q^{(0)} = T \sum_k \frac{k_x + a + q_x/2}{m} [\hat{G}_0]_{k+q, k}, \quad (26)$$

$$j_q^{(1)} = \sum_{q'} K^{(1)}(q, q') V_{q'}, \quad (27)$$

$$j_q^{(2)} = \sum_{q'q''} K^{(2)}(q, q', q'') V_{q'} V_{q''}. \quad (28)$$

The linear response function $K^{(1)}(q, q')$ can be identified with the non-interacting correlation function between density and current-density,

$$K^{(1)}(q, q') = T \sum_{kk'} \frac{k_x + a + q_x/2}{m} \times [\hat{G}_0]_{k+q, k'+q'} [\hat{G}_0]_{k', k}. \quad (29)$$

The quadratic response function is

$$K^{(2)}(q, q', q'') = T \sum_{kk'k''} \frac{k_x + a + q_x/2}{m} [\hat{G}_0]_{k+q, k'+q'} \times [\hat{G}_0]_{k', k''} [\hat{G}_0]_{k'' - q'', k}. \quad (30)$$

Graphical representations of $j_q^{(1)}$ and $j_q^{(2)}$ are shown in Figures 1a and b. It is instructive to see how functional averaging of these expressions with the effective action

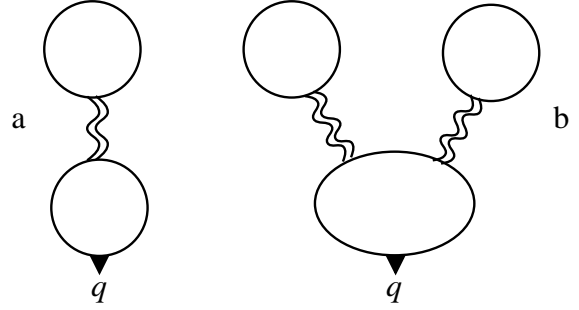


Fig. 2. Hartree contributions to the functional average of the non-equilibrium current density given in equations (23, 25). (a) Contribution from the linear response current $j_q^{(1)}$. (b) Contribution from the quadratic response $j_q^{(2)}$. The double wavy line is the RPA interaction.

(20) yields the well-known [13,14] interaction corrections to the equilibrium current to first order in the RPA interaction. Of course, the equilibrium current is more easily obtained from the derivative of the thermodynamic potential with respect to the static flux [13,14], but the following calculation clarifies the close connection between non-linear response and electron-electron interactions [4]. To perform the Gaussian integration, it is convenient to first eliminate the linear term in equation (20) by redefining the Φ -field such that its Gaussian average vanishes. This is achieved with the help of the shift-transformation $\tilde{\Phi}_q = \Phi_q - iT^{-1} \sum_{q'} f_{qq'}^{RPA} N_0(q')$, or equivalently for $V_q = iT\Phi_q$,

$$V_q = \tilde{V}_q + \sum_{q'} f_{qq'}^{RPA} N_0(q'). \quad (31)$$

Here $f_{qq'}^{RPA}$ is the *inverse* of the infinite matrix with elements $\delta_{qq'} f_{\mathbf{q}}^{-1} + \Pi_0(q, q')$. It follows that within the Gaussian approximation

$$\langle \tilde{V}_q \rangle_{S_{eff}} = 0, \quad (32)$$

$$\langle \tilde{V}_q \tilde{V}_{-q'} \rangle_{S_{eff}} = -T f_{qq'}^{RPA}. \quad (33)$$

Substituting equation (31) into equations (27, 28), and averaging over the \tilde{V} -field in Gaussian approximation, it is now easy to show that

$$\langle j_q^{(1)} \rangle_{S_{eff}} = \sum_{q'} \sum_{q_1} K^{(1)}(q, q') f_{q'q_1}^{RPA} N_0(q_1), \quad (34)$$

$$\langle j_q^{(2)} \rangle_{S_{eff}} = \sum_{q'q''} \sum_{q_1q_2} K^{(2)}(q, q', q'') f_{q'q_1}^{RPA} f_{q''q_2}^{RPA} \times N_0(q_1) N_0(q_2) - T \sum_{q'q''} K^{(2)}(q, q', -q'') f_{q'q''}^{RPA}. \quad (35)$$

Graphically equation (34) and the first term in equation (35) can be represented by the Hartree diagrams shown in Figure 2, while the second term in equation (35) is represented by the Fock diagram in Figure 3. To see that for $q = 0$ equations (34, 35) reduce to the well-known [13,14] first order (in the RPA interaction) corrections to

the equilibrium current, we use the following exact identity

$$\sum_{\mathbf{k}'} G_0(\mathbf{k}, \mathbf{k}', i\tilde{\omega}_n) \frac{k'_x + a}{m} G_0(\mathbf{k}', \mathbf{k}'', i\tilde{\omega}_n) = \frac{L\phi_0}{2\pi} \frac{\partial}{\partial\phi} G_0(\mathbf{k}, \mathbf{k}'', i\tilde{\omega}_n), \quad (36)$$

where $[\hat{G}_0]_{kk'} = \delta_{nn'} G_0(\mathbf{k}, \mathbf{k}', i\tilde{\omega}_n)$. Equation (36) can be easily proven by taking the derivative of both sides of equation (17) (with $V(\mathbf{r}, \tau)$ set equal to zero) with respect to a . With the help of this identity we obtain for the functional average of the linear response current (34)

$$\frac{-e}{L} \langle j_0^{(1)} \rangle_{S_{eff}} = -\frac{c}{2} \sum_{\mathbf{q}\mathbf{q}'} f_{\mathbf{q}\mathbf{q}'}^{RPA} \frac{\partial}{\partial\phi} [N_0(-\mathbf{q})N_0(\mathbf{q}')], \quad (37)$$

where $f_{\mathbf{q}\mathbf{q}'}^{RPA} \equiv f_{\mathbf{q}0, \mathbf{q}'0}^{RPA}$ is the static RPA interaction. Similarly, for $q = 0$, equation (35) can be written in the form

$$\frac{-e}{L} \langle j_0^{(2)} \rangle_{S_{eff}} = -\frac{c}{2} \sum_{\mathbf{q}\mathbf{q}'} \left[\frac{\partial}{\partial\phi} f_{\mathbf{q}\mathbf{q}'}^{RPA} \right] N_0(-\mathbf{q})N_0(\mathbf{q}') + I_F, \quad (38)$$

where

$$I_F = -\frac{c}{2} \sum_{\mathbf{q}\mathbf{q}'} T \sum_n f_{\mathbf{q}\mathbf{q}'}^{RPA} \frac{\partial}{\partial\phi} \Pi_0(\mathbf{q}, \mathbf{q}', i\omega_n) \quad (39)$$

is the Fock contribution to the equilibrium persistent current [13, 14]. The sum of equation (37) and the first term in equation (38) yield the (non self-consistent) Hartree contribution to the equilibrium current,

$$I_H = -\frac{c}{2} \frac{\partial}{\partial\phi} \sum_{\mathbf{q}\mathbf{q}'} f_{\mathbf{q}\mathbf{q}'}^{RPA} N_0(-\mathbf{q})N_0(-\mathbf{q}'). \quad (40)$$

We have argued elsewhere [15] that the neglect of self-consistency in equation (40) does not properly take into account the subtle interplay between disorder and interactions, so that the correct order of magnitude of the Hartree current can only be obtained by means of a self-consistent calculation.

3 Quadratic response to an external electric field

3.1 Derivation of the non-equilibrium current from the Fock correction to the equilibrium current

Our rather unconventional derivation of the interaction correction to the equilibrium current makes the connection between electron-electron interactions and non-linear response manifest. In fact, from our derivation it is clear that the Fock contribution (39) to the equilibrium current is closely related to the non-equilibrium current given in equation (1). Physically our Hubbard-Stratonovich field Φ

can be identified with the scalar potential of electromagnetism, which is generated self-consistently by the motion of the electrons [5, 8]. Therefore the negative gradient of our auxiliary potential $V(\mathbf{r}, \tau)$ is the effective force acting on the electrons, which in turn can be associated with an internal electric field $\mathbf{E}(\mathbf{r}, \tau)$,

$$-e\mathbf{E}(\mathbf{r}, \tau) = -\nabla V(\mathbf{r}, \tau). \quad (41)$$

Defining $\mathbf{E}(\mathbf{r}, \tau) = \sum_q e^{i(\mathbf{q}\cdot\mathbf{r} - \omega_n\tau)} \mathbf{E}_q$ and using equation (18), we have $e\mathbf{E}_q = i\mathbf{q}V_q$, or

$$V_q = -ie \frac{\hat{\mathbf{q}} \cdot \mathbf{E}_q}{|\mathbf{q}|}, \quad (42)$$

where $\hat{\mathbf{q}} = \mathbf{q}/|\mathbf{q}|$. From equation (33) we thus conclude

$$T f_{\mathbf{q}\mathbf{q}'}^{RPA} = -\frac{e^2}{|\mathbf{q}||\mathbf{q}'|} \langle (\hat{\mathbf{q}} \cdot \mathbf{E}_q) (\hat{\mathbf{q}}' \cdot \mathbf{E}_{-q'}) \rangle_{S_{eff}}, \quad (43)$$

so that the Fock contribution (39) to the equilibrium current can be written as

$$I_F = \sum_{\mathbf{q}\mathbf{q}'} \sigma^{(2)}(\mathbf{q}, \mathbf{q}', i\omega_n) \langle (\hat{\mathbf{q}} \cdot \mathbf{E}_q) (\hat{\mathbf{q}}' \cdot \mathbf{E}_{-q'}) \rangle_{S_{eff}}, \quad (44)$$

where the non-linear conductivity $\sigma^{(2)}(\mathbf{q}, \mathbf{q}', i\omega_n)$ is given by

$$\sigma^{(2)}(\mathbf{q}, \mathbf{q}', i\omega_n) = \frac{c}{2} \frac{e^2}{|\mathbf{q}||\mathbf{q}'|} \frac{\partial}{\partial\phi} \Pi_0(\mathbf{q}, \mathbf{q}', i\omega_n). \quad (45)$$

Note that functional averaging restores translational invariance in time, so that the average in equation (44) is proportional to $\delta_{nn'}$. This equation shows that the Fock current can be viewed as the sum of functionally averaged non-equilibrium currents, generated in second order in the internal electric fields associated with the motion of the electrons. Clearly, the corresponding non-linear response function $\sigma^{(2)}$ is a system property that should be independent of the origin of the electric fields. In particular, if we add an external electric field, equation (44) is still valid provided we identify \mathbf{E} with the total electric field. Thus, after performing in equation (45) the usual analytic continuation, $i\omega_n \rightarrow \omega + i0^+$, we conclude that the non-linear conductivity $\sigma^{(2)}(\omega, \phi)$ defined in equation (1) can be identified with

$$\sigma^{(2)}(\omega, \phi) = \lim_{\mathbf{q}, \mathbf{q}' \rightarrow 0} \sigma^{(2)}(\mathbf{q}, \mathbf{q}', \omega + i0^+). \quad (46)$$

Recall that $\sigma^{(2)}(\omega, \phi)$ describes the *time-independent* part of the non-equilibrium current that is proportional to the intensity of a *time-dependent*, spatially uniform electric field.

Two remarks are in order. The first concerns the analytic continuation of the imaginary frequency response to real frequencies. Because equation (45) relates a non-linear response function to the flux-derivative of a linear response function (the polarization), we may rely on the fluctuation-dissipation theorem to relate real and imaginary time response. Of course, equation (45) can also

be obtained with the help of the identity (36) from the expression (30) for the general non-linear response function $K^{(2)}(q, q', q'')$. It is not difficult to see that quite generally the correct real frequency response can be obtained by replacing $i\omega_n \rightarrow \omega + i0^+$, $i\omega_{n'} \rightarrow \omega' + i0^+$, and $i\omega_{n''} \rightarrow \omega'' + i0^+$ for all frequencies [12].

Secondly, we would like to emphasize that so far we have not averaged over the disorder potential, *i.e.* all equations given above are valid for an arbitrary realization of $U(\mathbf{r})$. In the following subsection we shall start from equation (46) to perform the disorder average of the non-linear conductivity. This has the advantage that we only need to average a product of *two* Green's functions. Note that in the work [4] the average of the non-linear conductivity has been calculated directly from equations (28, 30).

3.2 The relation between non-linear conductivity, polarization, and linear conductivity

Using the fact that $\overline{\Pi}_0(\mathbf{q}, \mathbf{q}', i\omega_n) = \delta_{\mathbf{q}\mathbf{q}'} \overline{\Pi}_0(\mathbf{q}, i\omega_n)$, we see from equations (44, 45) that the disorder averaged static non-equilibrium current that is generated at quadratic order in a space- and time-dependent longitudinal electric field with Fourier components $\mathbf{E}(\mathbf{q}, \omega)$ is given by

$$\overline{I}^{(2)} = \overline{\sigma}^{(2)}(\mathbf{q}, \omega, \phi) |\mathbf{E}(\mathbf{q}, \omega)|^2, \quad (47)$$

with

$$\overline{\sigma}^{(2)}(\mathbf{q}, \omega, \phi) = \frac{c e^2}{2 \mathbf{q}^2} \frac{\partial}{\partial \phi} \overline{\Pi}_0(\mathbf{q}, \omega). \quad (48)$$

We thus need to know the flux-dependent part of the disorder averaged polarization of the ring. For frequencies $|\omega| \lesssim \Delta$ and wave-vectors $|\mathbf{q}| \lesssim 2\pi/\ell$ (where $\ell = v_F \tau$ is the elastic mean free path) non-perturbative methods are necessary to calculate this quantity [16]. Here we are interested in the high frequency regime $|\omega| \gtrsim \Delta$, where we may use the impurity diagram technique [17]. However, for $|\omega| \tau \lesssim 1$ and $|\mathbf{q}| \ell \lesssim 1$ the direct diagrammatic calculation of $\frac{\partial}{\partial \phi} \overline{\Pi}_0(\mathbf{q}, \omega)$ is not so easy, because there exists non-trivial cancellations between vertex corrections to the density vertices [14, 18]. Physically, these corrections arise from the diffusive motion of the electrons in the disordered metal. To take these corrections into account without having to perform complicated manipulations, we use the exact relation between irreducible polarization and longitudinal conductivity $\overline{\sigma}(\mathbf{q}, \omega)$ [19], which in our normalization [10] reads

$$\overline{\Pi}(\mathbf{q}, \omega) = i \frac{\mathbf{q}^2 \mathcal{V}}{\omega e^2} \overline{\sigma}(\mathbf{q}, \omega). \quad (49)$$

From equation (48) we thus obtain

$$\overline{\sigma}^{(2)}(\mathbf{q}, \omega, \phi) = \frac{c}{2} \frac{\mathcal{V}}{(-i\omega)} \frac{\partial}{\partial \phi} \overline{\sigma}(\mathbf{q}, \omega). \quad (50)$$

At finite \mathbf{q} , the dynamic conductivity has a diffusion pole [19]. In fact, according to reference [20] in the limit of small wave-vectors and frequencies

$$\overline{\sigma}(\mathbf{q}, \omega) = \frac{i\omega}{i\omega - \mathcal{D}(\omega) \mathbf{q}^2} \overline{\sigma}(\omega), \quad (51)$$

where $\overline{\sigma}(\omega) = \overline{\sigma}(0, \omega)$, and the frequency-dependent diffusion coefficient $\mathcal{D}(\omega)$ is related to the dynamic conductivity *via* [20]

$$\frac{\mathcal{D}(\omega)}{\mathcal{D}_0} = \frac{\overline{\sigma}(\omega)}{\sigma_0}. \quad (52)$$

Here \mathcal{D}_0 is the classical diffusion coefficient, which is related to the Drude conductivity σ_0 *via* the Einstein relation $\mathcal{D}_0 = (\Delta \mathcal{V})^{-1} e^2 \sigma_0$. We thus conclude

$$\overline{\sigma}^{(2)}(\mathbf{q}, \omega, \phi) = \frac{c}{2} \mathcal{V} \frac{\partial}{\partial \phi} \left[\frac{\overline{\sigma}(\omega)}{\mathcal{D}(\omega) \mathbf{q}^2 - i\omega} \right]. \quad (53)$$

Diagrammatically, the diffusion pole in equations (51, 53) implicitly takes the so-called *diffusion* diagrams into account [17]. On the other hand, the weak-localization corrections described by the *Cooperon* diagrams have to be included explicitly in the calculation $\overline{\sigma}(\omega)$ and $\mathcal{D}(\omega)$. These diagrams are responsible for the dominant dependence on the magnetic flux.

3.3 Averaging over disorder

According to equations (52, 53) the average non-linear conductivity can be expressed in terms of the flux-dependent part of the average linear conductivity. The latter is determined by the famous weak localization correction arising from coherent backscattering [17],

$$\begin{aligned} \frac{\partial}{\partial \phi} \overline{\sigma}(\omega) &= -\frac{e^2 \mathcal{D}_0}{\pi \mathcal{V}} \\ &\times \frac{\partial}{\partial \phi} \sum'_{\mathbf{Q}} \frac{1}{\mathcal{D}_0(\mathbf{Q} + 2\mathbf{a})^2 - i\omega + \mathcal{D}_0/L_\phi^2}, \end{aligned} \quad (54)$$

where the prime means that the sum is restricted to $|\mathbf{Q}| \lesssim 2\pi/\ell$, and L_ϕ is the dephasing length [17]. Scaling out the Thouless energy $E_c = \mathcal{D}_0/L^2$ and setting now $\mathbf{q} = 0$ in equation (53), we obtain from equation (54)

$$\overline{\sigma}^{(2)}(\omega, \phi) = \frac{c E_c (eL/E_c)^2}{(-i\bar{\omega}) \phi_0} g(\omega, \phi), \quad (55)$$

where $\bar{\omega} = \omega/E_c$, and the dimensionless function $g(\omega, \phi)$ is given by

$$g(\omega, \phi) = -\frac{\phi_0}{2\pi} \frac{\partial}{\partial \phi} \sum'_{\mathbf{Q}} \frac{E_c}{\mathcal{D}_0(\mathbf{Q} + 2\mathbf{a})^2 - i\omega + \mathcal{D}_0/L_\phi^2}. \quad (56)$$

For a thin ring with $L_\perp \lesssim \ell$ the \mathbf{Q} -summation in equation (56) is one-dimensional, and can be carried out exactly, with the result

$$g(\omega, \phi) = \frac{2e^{-W}}{W} \frac{\sin(4\pi\phi/\phi_0)[1-e^{-2W}]}{[1-2e^{-W}\cos(4\pi\phi/\phi_0)+e^{-2W}]^2}. \quad (57)$$

Here $W = \sqrt{(L/L_\varphi)^2 - i\bar{\omega}}$, where the root has to be taken such that $\Re W \geq 0$. For $|W| \ll 1$ this reduces to

$$g(\omega, \phi) = \frac{4\sin(4\pi\phi/\phi_0)}{[4\sin^2(2\pi\phi/\phi_0) - i\bar{\omega} + (L/L_\varphi)^2]^2}. \quad (58)$$

Note that by definition $L_\varphi \gg L$ in a mesoscopic system, so that for $|\omega| \ll E_c$ the parameter $|W|$ is smaller than unity. On the other hand, for $|\omega| \gtrsim E_c$ the prefactor e^{-W} in equation (57) reduces to $\exp[-\sqrt{|\bar{\omega}|/2}]$, so that the non-linear conductivity becomes exponentially small. We disagree in this point with Kravtsov and Yudson [4], who found that the non-linear conductivity in the regime $E_c \ll \omega \ll \tau^{-1}$ is finite and approximately frequency-independent. In view of the close connection between the non-linear conductivity and the Fock contribution to the equilibrium persistent current discussed above, we think that our result is physically more reasonable. The fact that for $|\omega| \gg E_c$ the non-linear conductivity (55) is exponentially small is closely related to the exponential suppression of the contribution from Matsubara frequencies larger than E_c to the average Fock current \bar{I}_F in equation (39).

4 Possible experimental tests

For simplicity, let us consider a time-dependent but spatially constant external electric field along the circumference of the ring,

$$E(t) = E(\omega) \cos(\omega t). \quad (59)$$

Because at zero wave-vector and finite frequencies the polarization $\bar{\Pi}(0, \omega)$ vanishes, this field is not screened. Hence, $\hat{\mathbf{q}} \cdot \mathbf{E}(\mathbf{q} = 0, \omega)$ in equation (47) can be identified with the external field $E(\omega)$. Experimentally, the field (59) can be generated by a time-dependent magnetic flux through the center of the ring,

$$\phi(t) = \phi + \phi(\omega) \sin(\omega t). \quad (60)$$

By Faraday's law of induction, the relation between $E(\omega)$ and $\phi(\omega)$ is

$$eLE(\omega) = 2\pi\omega \frac{\phi(\omega)}{\phi_0}. \quad (61)$$

Note that in the experiment [2] the current was measured in the presence of such a time-dependent flux with frequencies in the range between 10 and 10^3 Hz. In this range no frequency-dependence of the current was detected, so that apparently the measurements were performed in the static limit. Note, however, that in principle one should

distinguish between the thermodynamic equilibrium current that is determined by the flux-dependent part of the free energy, and the dynamic current that is obtained from the time-dependent response in the limit of vanishing frequency [21]. In the present work we are interested in the frequency range $\Delta \ll \omega \ll \tau^{-1}$, corresponding to frequencies between 10^8 and 10^{13} Hz. We predict that the static non-equilibrium current should become exponentially small as soon as the frequency of the electric field exceeds the Thouless energy E_c . *This effect can be used to directly measure the Thouless energy of a mesoscopic ring.* For the rings used in reference [2] the time-dependent non-equilibrium current should disappear for $\omega \approx 10^{10}$ Hz. We would like to emphasize that this prediction can be verified without any modifications of the experimental setup used in references [2, 3].

Because the non-equilibrium current $\bar{I}^{(2)}$ is driven by an external time-dependent flux $\phi(\omega)$, it can be easily distinguished from the thermodynamic equilibrium current. Let us now discuss the expected size of this non-equilibrium current. Given the fact that the external field in equation (59) has a cos-dependence, the experimentally measured non-equilibrium current is determined by the real part of the non-linear conductivity,

$$\bar{I}^{(2)} = \Re [\bar{\sigma}^{(2)}(\omega, \phi)] \left[\frac{\omega}{eL} \right]^2 \left[\frac{2\pi\phi(\omega)}{\phi_0} \right]^2. \quad (62)$$

For frequencies $|\omega| \lesssim E_c$ we may use equation (58) to simplify the non-linear conductivity, so that in this regime we obtain after some rescalings

$$\bar{I}^{(2)} \approx -\frac{cE_c}{\phi_0} \left[\frac{2\pi\phi(\omega)}{\phi_0} \right]^2 |\bar{\omega}|^{-1/2} f(\bar{\phi}, \bar{\omega}, \gamma), \quad (63)$$

where $\bar{\phi} = \phi/\phi_0$, $\bar{\omega} = \omega/E_c$, $\gamma = \mathcal{D}_0/(L_\varphi^2|\omega|)$. Defining the dimensionless variable

$$X = 2 \frac{\sin(2\pi\bar{\phi})}{\sqrt{|\bar{\omega}|}}, \quad (64)$$

the function $f(\bar{\phi}, \bar{\omega}, \gamma)$ can be written as

$$f(\bar{\phi}, \bar{\omega}, \gamma) = 8 \cos(2\pi\bar{\phi}) \frac{X[\gamma + X^2]}{[1 + [\gamma + X^2]^2]^2}. \quad (65)$$

A graph of $f(\bar{\phi}, \bar{\omega}, \gamma)$ as function of $\bar{\phi} = \phi/\phi_0$ for $\bar{\omega} = 0.1$ and $\gamma = 1$ is shown in Figure 4. Obviously the size of the current $\bar{I}^{(2)}$ in equation (63) is determined by three experimentally controllable parameters: $\phi(\omega)$, ϕ , and ω . Let us find the values of these parameters that maximize the current. Obviously $\phi(\omega)$ should be chosen as large as possible. It should be kept in mind, however, that equation (63) is the quadratic order in a systematic expansion in powers of $\phi(\omega)/\phi_0$. Higher orders should be negligible as long as $|\phi(\omega)| \ll \phi_0$. Thus, the largest value of $|\phi(\omega)|$ where equation (63) can be expected to be accurate is

$$|\phi(\omega)| \approx \frac{\phi_0}{2\pi}. \quad (66)$$

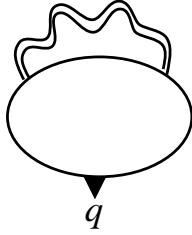


Fig. 3. Fock contribution to functional average of $j^{(2)}$.

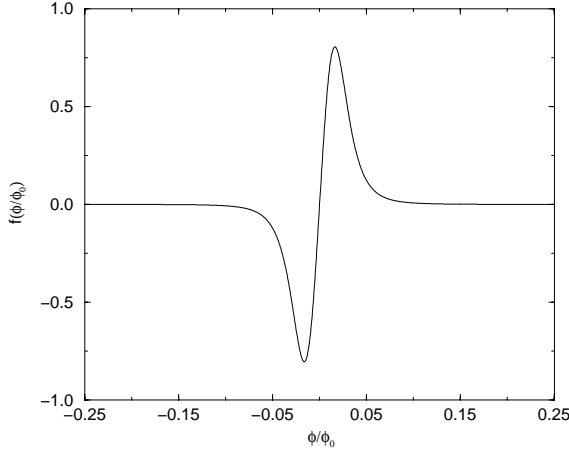


Fig. 4. $f(\bar{\phi}, \bar{\omega}, \gamma)$ as function of ϕ/ϕ_0 for $\bar{\omega} = 0.1$ and $\gamma = 1$.

Next, consider the optimal choice of the frequency. Because of the factor of $|\bar{\omega}|^{-1/2}$ in equation (63), it is advantageous to choose the frequency as small as possible. However, our perturbative calculation breaks down for frequencies of the order of the mean level spacing Δ . The optimal choice is therefore

$$\omega \approx \Delta. \quad (67)$$

Finally, from equation (65) and Figures 5, 6 it is clear that the function $\tilde{f}(X, \gamma) \equiv f/\cos(2\pi\bar{\phi})$ has a maximum at a value $X_m(\gamma) = O(1)$ (provided γ is not too large). This implies that for $|\omega| \ll E_c$ the non-equilibrium current is maximal at the static flux $\phi = \phi_m$, where

$$\phi_m = \frac{\phi_0}{4\pi} \sqrt{\frac{|\omega|}{E_c}} X_m \left(\frac{D}{L_\varphi^2 |\omega|} \right). \quad (68)$$

The function $X_m(\gamma)$ is shown in Figure 6. From equation (64) we also see that the width $\Delta\phi$ of the interval around ϕ_m (modulo $\phi_0/2$) where the current is enhanced is $\Delta\phi \approx \phi_0 |\bar{\omega}|^{1/2}/(4\pi)$. Outside this interval, which is rather narrow for $|\bar{\omega}| \ll 1$, the non-equilibrium current is much smaller than at the maxima (see Fig. 4). In fact, for $|\phi - \phi_m| \gg \Delta\phi$ the parameter $|X|$ in equation (64) is large compared with unity, so that we may approximate

$$f(\bar{\phi}, \bar{\omega}, \gamma) \approx \frac{|\bar{\omega}|^{5/2} \cos(2\pi\bar{\phi})}{4 \sin^5(2\pi\bar{\phi})}, \quad |X| \gg 1, \quad (69)$$

where we have assumed that $\gamma \lesssim 1$. Using $2 \sin^2(x) = [1 - \cos(2x)]$, the non-equilibrium current in this regime can

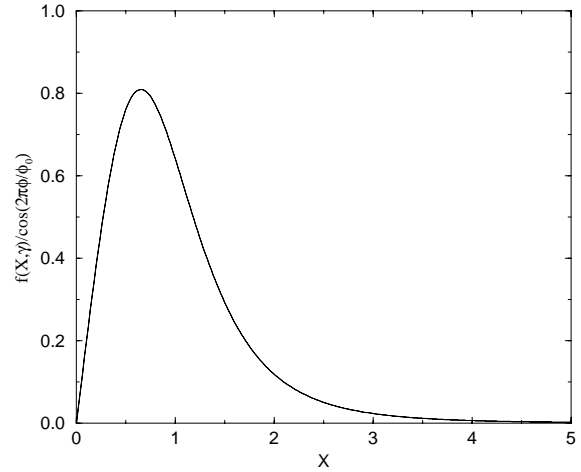


Fig. 5. $\tilde{f}(X, \gamma) = f/\cos(2\pi\bar{\phi})$ as function of X (see Eq. (64)) for $\gamma = 1$.

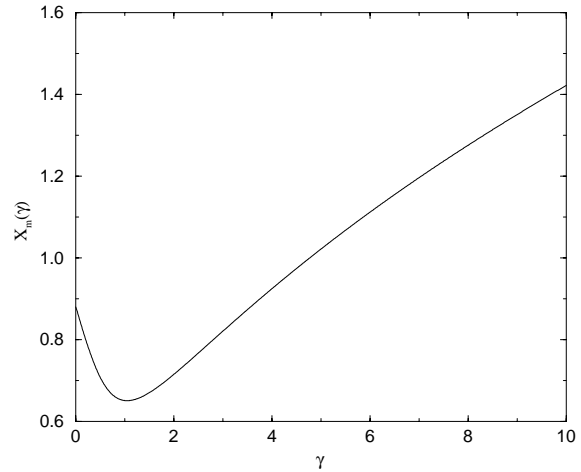


Fig. 6. Position of the maximum $X_m(\gamma)$ of $\tilde{f}(X, \gamma)$ as function of γ . For $\gamma = 0$ it is easy to show that $X_m(0) = (3/5)^{1/4} \approx 0.88$, while $X_m(\gamma) \propto \sqrt{\gamma}$ for $\gamma \rightarrow \infty$.

be written as

$$\bar{I}^{(2)} \approx -\frac{cE_c}{\phi_0} \left[\frac{2\pi\phi(\omega)}{\phi_0} \right]^2 |\bar{\omega}|^2 \frac{\sin(4\pi\bar{\phi})}{[1 - \cos(4\pi\bar{\phi})]^3}. \quad (70)$$

For the parameters of the experiment [2] (taking now the spin degeneracy into account), we find that at the optimal values of the parameters given in equations (66–68) the maximal amplitude of the non-equilibrium current is $\bar{I}_{max}^{(2)} \approx 5 \times 10^{-3} e v_F / L$. This current has the same order of magnitude as the equilibrium current measured in references [2, 3], and therefore should be measurable with the available technology. The rather pronounced peaks of $\bar{I}^{(2)}$ as function of ϕ for small values of ϕ (modulo $\phi_0/2$) distinguish the non-equilibrium current from the thermodynamic persistent current.

5 Summary

In this paper we have presented a thorough theoretical analysis of the static non-equilibrium current $I^{(2)}$ in a mesoscopic metal ring threaded by a magnetic flux, which is generated in quadratic response to a time-dependent electric field. Using a path integral approach, we have shown that this non-equilibrium current is closely related to the screened exchange contribution to the thermodynamic persistent current [13,14]. In fact, from equations (39, 44, 45) it is obvious that the weight of the RPA screened interaction in the exchange correction to the equilibrium persistent current at fixed energy-momentum transfer is essentially given by the non-linear conductivity $\sigma^{(2)}(\omega, \phi)$ associated with $I^{(2)}$. This observation has allowed us to derive the relation (48) between the non-linear conductivity and the flux-dependent part of the polarization. Thus, the disorder average $\bar{\sigma}^{(2)}(\omega, \phi)$ of the non-linear conductivity is directly related to the weak-localization correction to the average polarization, which in turn can be expressed in terms of the weak-localization correction to the linear conductivity. In other words, *the average non-linear conductivity is directly related to the weak localization correction to the linear conductivity.*

A measurement of the average static non-equilibrium current $\bar{I}^{(2)}$ in a mesoscopic metal ring as function of frequency and static flux would be very interesting for several reasons. Such a measurement would directly probe the weak localization corrections to the frequency-dependent polarization and the linear conductivity of the system. In contrast to Kravtsov and Yudson [4], we predict that the static non-equilibrium current should be exponentially suppressed when the external frequency exceeds the Thouless energy [22]. This is in agreement with the close connection between the non-equilibrium current $I^{(2)}$ and the Fock contribution I_F to the equilibrium persistent current. The latter is known to be exponentially suppressed if the temperature becomes larger than the Thouless energy [13]. Note that in this case the temperature acts as an infrared cutoff, just like the external frequency in the case of the non-equilibrium current. Our theory can be verified experimentally from the measurement of the *time-independent* non-linear current response of a mesoscopic metal ring pierced by a *time-dependent* external flux $\phi(t)$ of the form (60), with frequencies in the range $\Delta \lesssim \omega \ll \tau^{-1}$. We hope that such an experiment will be done in the near future.

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