Dynamic thermoelectric and heat transport in mesoscopic capacitors

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We discuss the low-frequency response of charge and heat transport to oscillatory voltage and temperature shifts in mesoscopic capacitors. We obtain within scattering theory generic expressions for the quantum admittances up to second order in the ac frequencies in terms of electric, thermoelectric and heat capacitances and relaxation resistances. Remarkably, we find that the thermocurrent can lead or lag the applied temperature depending on the gate voltage applied to a quantum RC circuit. Furthermore, the relaxation resistance for cross terms becomes nonuniversal as opposed to the purely electric or thermal cases.

Introduction. Time-dependent charge transport in quantum conductors subject to ac electric fields provides insight into electronic dynamics at nanoscale dimensions. For low frequencies the dynamics of a mesoscopic conductor is characterized by the \( R_G C_G \) time, where \( C_G \) is the quantum capacitance and \( R_G \) is the charge relaxation resistance. Whereas the former generally depends on energy, the latter unexpectedly attains the constant value of \( h/2e^2 \) in a single-channel quantum capacitor. These predictions are confirmed by ac measurements in mesoscopic RC circuits. Further developments have led to the experimental demonstration of coherent single-electron emitters. This achievement has spurred an enormous interest in both the fundamentals of time-resolved electronic transport (both experimentally and theoretically) and its applications to, e.g., metrology and quantum information processing, just to mention a few.

Electronic current, however, can also be driven by thermal gradients. In the stationary case the Seebeck effect leads to the generation of thermovoltages in response to applied temperature differences in open circuits. While dc thermopower has been extensively investigated in nanostructures, the ac Seebeck effect has received little attention to date. The subject is interesting for several reasons. First, treating voltage and thermal driving fields on an equal footing opens up the door to not only electrical but also thermodynamic characterizations of mesoscopic systems. In fact, ac calorimetry techniques have been successfully applied to superconducting loops. What can be learned from an analogous experiment with normal conductors? Second, quantum refrigeration devices based on the Peltier effect (reciprocal to Seebeck) typically use static currents. What can we expect in the ac regime of transport? Third, how does Coulomb interaction renormalize the RC parameters in a thermoelectric device? These are the kind of questions we want to address in this work.

Consider a multichannel mesoscopic conductor coupled to a single terminal as in Fig. 1(a). The sample is driven out of equilibrium with oscillating voltages \( \delta V(\omega) \) and temperatures \( \delta T(\omega) \) applied to the reservoir. The driving fields operate with regard to an equilibrium state described by the chemical potential \( \mu \) and the base temperature \( T_0 \). Hence, the linear-response electric \( \delta I \) and heat \( \delta J \) currents are given by

\[
\begin{pmatrix}
\delta I \\
\delta J
\end{pmatrix} =
\begin{pmatrix}
G(\omega) & L(\omega) \\
M(\omega) & K(\omega)
\end{pmatrix}
\begin{pmatrix}
\delta V \\
\delta T
\end{pmatrix},
\]

where the \( 2 \times 2 \) Onsager matrix includes diagonal elements [electric \( G(\omega) \) and thermal \( K(\omega) \) admittances] and nondiagonal coefficients [thermoelectric \( L(\omega) \) and electrothermal \( M(\omega) \) admittances]. The latter are related by reciprocity.

In a low-frequency expansion, \( G(\omega) = -i\omega C_G + \omega^2 C_G^2 R_G \), the imaginary part of the electric admittance

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\[
\frac{1}{2}
\begin{pmatrix}
\Re & \Im \\
-\Im & \Re
\end{pmatrix}
\begin{pmatrix}
\delta V \\
\delta T
\end{pmatrix},
\]

where \( \Re \) and \( \Im \) denote real and imaginary parts of the Onsager matrix. For example, the heat admittance \( M(\omega) \) can be expressed in terms of the electric admittance \( G(\omega) \) and its derivative with respect to temperature

\[
M(\omega) = \frac{\partial G(\omega)}{\partial T},
\]

with \( \partial G(\omega) \) divided by the Onsager matrix element \( K(\omega) \).
provides information on the electric capacitance or emittance of the system whereas its real part is directly related to the dissipation in the conductor. When transport is phase coherent, $R_G = \hbar/2e^2$ takes an universal value independently on the transmission value and it scales with $N$ the number of propagating channels [1]. Below, we demonstrate that the low temperature thermal relaxation resistance also becomes quantized but, surprisingly, its thermoelectric analog is always nonuniversal. Importantly, our gauge-invariant theory also includes the effect of Coulomb interactions taking into account the charges that pile-up when the sample is driven out of equilibrium by oscillatory voltage and thermal biases. We will illustrate our ac thermoelectromagnetic scattering theory with an application to a quantum capacitor system.

Scattering formalism. We present a scattering approach for coupled charge and energy transport. In general, the transport properties of a multichannel mesoscopic capacitor are described by the scattering matrix $S$ with elements $s_{nm}[E, U(\vec{r})]$ that relate the outgoing current amplitude in channel $n$ to the incoming current amplitude in channel $m$ for a carrier with a given energy $E$. The matrix is also a function of the internal potential landscape $U(\vec{r})$ built up inside the conductor. Quite generally, $U(\vec{r})$ is a function of the applied electrical and thermal biases [2]. Let us for the moment focus on the noninteracting case. Then, the time-dependent current and heat current operators are given by $\hat{I} = (e/\hbar) \int dEdE' \exp[i(E - E')t/\hbar][a^\dagger(E)a(E') - b^\dagger(E)b(E')]$ and $\hat{J} = (1/2\hbar) \int dEdE'(E + E' - 2\mu) \exp[i(E - E')t/\hbar][a^\dagger(E)a(E') - b^\dagger(E)b(E')]$ where $a = (a_1, a_2, \cdots, a_N)^\dagger$ and $b = (b_1, b_2, \cdots, b_N)^\dagger$ denote incoming and outgoing annihilation operators in the lead with $N$ channels and fulfill $b_n(E) = \sum_m s_{nm}(E)a_m(E)$.

We hereafter take $\mu \approx E_F$ (the Fermi energy) independently of $T_0$, which is a good approximation at low temperature, the regime of our interest.

The linear response of the electrical and heat currents to the ac external perturbations to the ac external perturbations in Eq. (1) is expressed as $G(\omega) = \delta I(\omega)/\delta V(\omega)$, $L(\omega) = \delta I(\omega)/\delta T(\omega)$, $M(\omega) = \delta J(\omega)/\delta V(\omega)$, and $K(\omega) = \delta J(\omega)/\delta T(\omega)$, where $I = \langle \hat{I} \rangle$ and $J = \langle \hat{J} \rangle$. The admittances can be obtained from the Kubo formulas [30, 32],

\begin{align*}
G(\omega) &= \frac{1}{\hbar \omega} \int_0^\infty dt \ e^{i(\omega + \delta \omega^\ast)t} \langle [\hat{I}(t), \hat{I}(0)] \rangle, \quad (2a) \\
K(\omega) &= \frac{1}{\hbar \omega T_0} \int_0^\infty dt \ e^{i(\omega + \delta \omega^\ast)t} \langle [\hat{J}(t), \hat{J}(0)] \rangle, \quad (2b) \\
L(\omega) &= \frac{1}{\hbar \omega T_0} \int_0^\infty dt \ e^{i(\omega + \delta \omega^\ast)t} \langle [\hat{I}(t), \hat{J}(0)] \rangle, \quad (2c) \\
M(\omega) &= \frac{1}{\hbar \omega} \int_0^\infty dt \ e^{i(\omega + \delta \omega^\ast)t} \langle [\hat{J}(t), \hat{I}(0)] \rangle. \quad (2d)
\end{align*}

By inserting the above expressions for $\hat{I}$ and $\hat{J}$ in Eq. (2a), we find the reciprocity relation $M(\omega) = T_0L(\omega)$, as expected. In the presence of an external magnetic field $B$, reciprocity becomes $M(\omega, B) = T_0L(\omega, -B)$. An important remark is now in order. Concerns have been recently raised about the validity of the fluctuation-dissipation theorem applied to heat transport [33]. In fact, Refs. [33, 34] find a nonvanishing term for the equilibrium heat-heat correlation function at $T_0 \to 0$, which is incompatible with the expected behavior of $K(\omega)$. However, this term is associated to scattering events that connect two different terminals and can therefore be safely ignored in our single-lead quantum capacitor system.

Inserting the charge and current operator expressions in Eq. (2) we find

\begin{align*}
G(\omega) &= \frac{e^2}{\hbar} \int dE \ Tr \{A(E, E + \hbar \omega)\hat{F}(E, \omega)\}, \quad (3a) \\
K(\omega) &= \frac{1}{\hbar T_0} \int dE \ E^2 \ Tr \{A(E, E + \hbar \omega)\hat{F}(E, \omega)\}, \quad (3b) \\
L(\omega) &= \frac{e}{\hbar T_0} \int dE \ E \ Tr \{A(E, E + \hbar \omega)\hat{F}(E, \omega)\}, \quad (3c)
\end{align*}

where the trace is over the transverse channels, $E_\nu = (E + \hbar \omega/2 - E_F)$, $A(E, E + \hbar \omega) = 1 - S^{(1)}(E)S(E + \hbar \omega)$, and $\hat{F}(E, \omega) = \{[\hat{f}_0(E) - \hat{f}_0(E + \hbar \omega)]/\hbar \omega \}$ with $\hat{f}_0 = 1/\{1 + \exp[(E - E_F)/k_BT_0]\}$ the Fermi function of the equilibrium reference state.

Low frequency analysis. The quantum capacitor exhibits pure ac response. Then, we expand Eq. (3) in powers of $\omega$. The leading-order contributions have the following functional form for $A = G$, $K$ and $L$:

\begin{align*}
A &= -i\omega C_A + \omega^2 C_A^2 R_A + \mathcal{O}(\omega^3), \quad (4)
\end{align*}

and

\begin{align*}
C_A &= g_A \int dE \left( \frac{\partial \hat{f}_0}{\partial E} \right) (E - E_F)^\lambda \rho(E), \quad (5)
\end{align*}

where $g_A$ and $r_A$ are constants to be specified below.

In Eq. (5) we define $\rho(E) = Tr[S^\dagger \partial_E S]/2\pi i$ as the density of states for the capacitor plate [8]. Notice that our formalism considers scattering states only. Thus, contributions to $\rho(E)$ due to bound states are not included. Furthermore, the index $\lambda$ in Eqs. (5) and (6) denotes the type of transport: $\lambda = 0$ (electric), $\lambda = 1$ (thermoelectric) or $\lambda = 2$ (thermal). We now discuss their main properties.

Electric admittance. For $A = G$ we have $g_G = e^2$.

In this case, $C_G$ is a quantum capacitance given by the density of states and can be accessed experimentally [33]. Equations (5) and (6) are generally valid for an arbitrary...
the dot, see Fig. 1(a). Then, the scattering amplitude phase that an electron picks up after a single turn around the potential curvature and where Ω is a characteristic energy scale of the potential field is applied perpendicular to the dot and propagating mode in the point contact. A strong magnification probability

We model the mesoscopic capacitor as a quantum \( N = 1 \) case because it is the experimentally relevant situation. We take the same parameters as Fig. 2.

FIG. 2: (a) Charge and (b) thermoelectric capacitances as a function of the gate voltage for the quantum system depicted in Fig. 1. Parameters: \( \epsilon = \Delta, \Omega = 3\Delta, \) and its denominator as

\[ G_{th} = \pi^{2}k_{B}^{2}T_{0}/3h \]

is the thermal conductance quantum, recently measured in a suspended nanostructure \[ 26 \]. Hence, \( R_{K} = 1/(2G_{th}) \) is the associated thermal relaxation resistance for a single mode mesoscopic capacitor. Equation (7) is also universal at low temperature. We note in passing that the characteristic time \( \tau_{K} = R_{K}C_{K} = h\rho(E_{F})/2 \) is always positive, as it should be, and equals the \( RC \) time for pure electric transport, \( \tau_{L} = R_{L}C_{L} = h\rho(E_{F})/2 \) \[ 1 \]. Furthermore, Eq. (7) scales as \( 1/N \) with the number of conducting channels, as occurs for the electrical case, because the numerator of Eq. (9) goes as \( N \) and its denominator as \( N^{2} \).

Thermoelectric admittance. Coupled charge and energy transport is governed at low frequencies by \( L(\omega) = -i\omega C_{L} + \omega^{2}C_{L}^{2}R_{L} \) with \( g_{L} = e^{2}/T_{0} \) and \( r_{L} = hT_{0}/2e \) in Eqs. (5) and (6) for \( \lambda = 1 \). At low temperature the thermoelectric capacitance becomes

\[ C_{L} = \frac{\pi^{2}}{3}ek_{B}^{2}T_{0}\rho(E_{F}) \]  

Remarkably, \( C_{L} \) can take positive or negative values for a conductor with a density of states that strongly depends on energy (e.g., a quantum dot with quasi-localized levels). This is in sharp contrast with the charge capacitance, which takes positive values only, as can be seen from its low-\( T_{0} \) expansion: \( C_{G} = e^{2}\rho(E_{F}) \). As a consequence, while electric currents lead \( \delta V(\omega) \) by \( \pi/2 \) in a capacitor, the thermocurrents lead or lag \( 6T \) depending on the sign of \( C_{L} \). This observation is confirmed for our dot capacitor, in which we observe periodic changes of sign whenever \( E_{F} - \epsilon_{0} \) crosses an energy level in the dot, see Fig. 2(b).
In the Seebeck effect, a voltage is generated in response to a temperature shift under the condition $I = 0$. Thus, the Seebeck coefficient is at linear response $S = -L/G$. We find in the low frequency limit that $S$ is given by a ratio of capacitances: $S(\omega) = -C_L/C_G + O(\omega)$. From our results in Fig. 2 we expect a strong dependence of $S$ as a function of energy in a mesoscopic capacitor. At low temperature, we find the Mott formula $S = (\pi^2k_B^2T_0/3e)\partial E\ln \rho(E)|_{E=E_F}$.

Note that the same energy dependence is obtained from a study of the Peltier effect since the Peltier coefficient is simply $\Pi = T_0S$ by reciprocity.

We find for the thermoelectric relaxation resistance $R_L$ at low temperature:

$$R_L = \frac{3h}{e\pi^2k_BT_0} \rho'(E)|_{E=E_F},$$

which again scales as $1/N$ for a $N$-channel conductor. Unlike the charge relaxation resistance and $R_K$ in the limit $T_0 \to 0$, the resistance $R_L$ is always a nonuniversal function that depends on the sample details. Surprisingly enough, we can recast the inverse of Eq. (9) in a more familiar form since $G_L \equiv R_L^{-1} = eG_{th}\partial E\ln \rho(E)|_{E=E_F}$ resembles the Mott formula for the dc thermopower and but applied to a seemingly unrelated quantity. In Fig. 3 we plot $R_L$ for the mesoscopic capacitor as a function of the gate voltage. As expected, the thermoelectric relaxation resistance never reaches a constant value. Note that the divergent behavior of $R_L$ in Fig. 3 occurs whenever the $\rho'(E_F)$ vanishes. Obviously the measurable quantity $\tau_L = R_L C_L = h\rho(E_F)$ is always finite and nonzero. Interestingly, the time scale for cross charge-energy transport is two times larger than purely charge or heat characteristic times. Then, for frequencies around $\tau_G^{-1}$, $\tau_{L}^{-1}$ or $\tau_{K}^{-1}$ (all of the same order) the $\omega^2$ term becomes comparable to the linear-in-$\omega$ term, allowing us to probe the characteristic times. For a quantum capacitor, we approximate $\rho(E_F) \sim 1/\Delta$ and estimate $\omega \sim 1$ GHz for a typical value $\Delta = 10$ meV. This frequency is not far from the scope of present technology for both voltage and temperature biases.

**Interactions.** A real calculation of admittance responses requires knowledge of the charge distribution inside the sample when the conductor is driven out of equilibrium by ac signals. Thus, we need to discuss screening effects. As a first approximation, we treat interactions within a mean-field theory.

Let $\delta U_0$ be the internal potential in the conductor away from its equilibrium value. For definiteness, we assume that $\delta U_0$ is spatially homogeneous [see Fig. 1(a) for an example]. The extension to inhomogeneous fields is straightforward. In the presence of interactions, the linear-response current becomes

$$\delta I = G(\omega)\delta V + L(\omega)\delta T + \Pi(\omega)\delta U_0,$$

with $\Pi(\omega)$ the screening function. Because $\delta I$ is invariant under a global voltage shift, $\Pi(\omega) = -G(\omega)$. On the other hand, the conductor is coupled to nearby gates via capacitance couplings. In the homogeneous case, we consider a single electric capacitance $C$. Then, the ac current is $\delta I = -i\omega C\delta U_0$ and using current conservation in Eq. (10) we find

$$\delta U_0 = \frac{G(\omega)\delta V + L(\omega)\delta T}{G(\omega) - i\omega C}.$$

We emphasize that the internal potential changes in response not only to voltage variations but also to temperature shifts.

Substituting Eq. (11) into Eq. (10) we obtain the electric and thermoelectric impedances, $G^{-1}(\omega) = G^{-1}(\omega) + i/\omega C$ and $L^{-1}(\omega) = L^{-1}(\omega) + iG(\omega)/\omega CL(\omega)$, respectively. Similarly, the interacting ac responses for the heat flow to oscillatory temperatures and voltages are $K(\omega) = K(\omega) - L(\omega)M(\omega)/[G(\omega) - i\omega C]$ and $\mathcal{M}(\omega) = M(\omega) - G(\omega)M(\omega)/[G(\omega) - i\omega C]$, with the latter obeying reciprocity ($\mathcal{M} = T_0\mathcal{C}$). Importantly, the four admittances constitute a gauge-invariant theory. Previous expressions can be further simplified in the low frequency limit. We find that the thermoelectric response $\mathcal{E}(\omega) = (-i\omega C_L + \omega^2C_L^\prime R_L)$ is expressed in terms of a renormalized thermoelectric capacitance $C_L^\prime = C_L C_G^2/C_G$, where the electrochemical capacitance $C_G^\prime$ is given by the geometrical and quantum capacitances in series: $(C_G^\prime)^{-1} = C^{-1} + C_G^{-1}$. Finally, the interacting heat admittance reads $K(\omega) = -\omega C_K + \omega^2(C_K^2 R_K - (C_L R_L + C_M R_M)C_L C_M/(C + C_G))$, where the renormalized heat capacitance is $C_K^\prime = C_K - C_M C_L/C$.

**Conclusion.** We have presented a scattering theory for the dynamical response of heat and electrical currents to oscillatory electrical and thermal signals applied to a mesoscopic capacitor. Our model includes interactions and focuses on the low-frequency properties of charge and energy transport. Importantly, we have found that the off-diagonal admittance matrix elements show crucial differences with the pure electric or thermal conductances: positive or negative phase delays and sample-dependent relaxation resistances. Our results are relevant both for thermodynamic characterizations of nanosystems and for prospect applications of thermoelectric nanodevices operating in the time domain.

**Acknowledgements.** We thank M. Büttiker and J. Splettstoesser for useful comments. This work was supported by MINECO Grants No. FIS2011-23526 and No. CSD2007-00042 (CPAN).

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