

Probing the energy reactance with adiabatically driven quantum dots

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The tunneling Hamiltonian describes a particle transfer from one region to the other. While there is no particle storage in the tunneling region itself, it has associated certain amount of energy. We name the corresponding flux *energy reactance* since, like an electrical reactance, it manifests itself in time-dependent transport only. Noticeably, this quantity is crucial to reproduce the universal charge relaxation resistance for a single-channel quantum capacitor at low temperatures. We show that a conceptually simple experiment is capable of demonstrating the existence of the energy reactance.

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Motivation. A very exciting experimental activity is lately taking place in search of controlling on-demand quantum coherent charge transport in the time domain. The recent burst of activity started with the experimental realization of quantum capacitors in quantum dots under ac driving [1], single particle emitters [2], and was followed by the generation of quantum charged solitons over the Fermi sea (levitons) [3]. A controlled manipulation of flying single electrons [4–6] and their time-resolved detection [7] have already been reported [8]. These marvelous developments, along with the identically impressive progress in the field of fast thermometry [9–11], are opening an avenue towards the study and control of the concomitant time-dependent energy flow in the quantum realm.

The relevant systems are characterized by small (nanoscale) components confining a small number of particles in contact to macroscopic reservoirs. This puts the description of the energy transport and heat generation beyond the scope of usual thermodynamical approaches, motivating a number of formal theoretical developments in statistical mechanics [12] and condensed matter physics [13]. At the heart of this problem, there is the proper definition of the quantum heat current in the time domain. The concept of heat looks very intuitive and anyone can provide a definition for it. Formally, it is a clear and well established concept in macroscopic systems close to equilibrium. However, its accurate definition at the nanoscale and in situations away from equilibrium is a deep and subtle issue, in particular due to the coupling between a nanosystem and macroscopic reservoirs; see, e.g., Refs. 14–23. In fact, while charge and energy are concepts obeying strict fundamental conservation laws, the definition of heat implies the proper identification of a portion of the total energy.

An appealing scenario to address this problem from

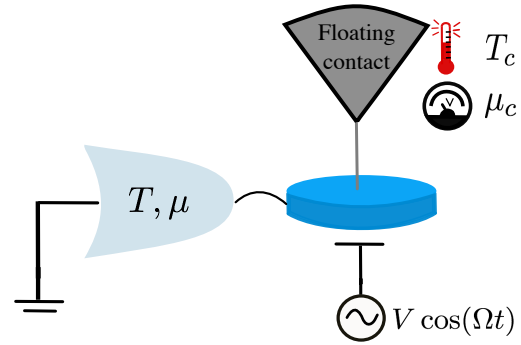


FIG. 1. Schematic of our proposal. The quantum RC circuit consists of a quantum dot (blue disk) coupled to a fermionic reservoir (light blue region) with well defined temperature T and chemical potential μ . Electrons can be transferred between the dot and the reservoir (black curve). The dot is capacitively coupled to a gate terminal where an ac potential of amplitude V and frequency Ω is applied. A floating contact is also attached to the dot. The temperature T_c and chemical potential μ_c of the floating contact adjust themselves to cancel both the charge and heat current flowing through it, thus allowing for an experimental test of the energy reactance, namely, the variation of the stored energy at the tunneling region between the floating contact and the dot (gray line).

the theoretical point of view is a periodically driven single level in contact to an electron reservoir. This is the most basic and meaningful setup to analyze the interplay of charge and energy dynamics. At the same time, this is the simplest model for a quantum RC circuit [24], which has been experimentally realized [1]. A sketch is presented in Fig. 1, where we stress that the driven level represents a quantum dot. The nonequilibrium ingredient is provided by the time-dependent gate voltage

$V(t) = V \cos(\Omega t)$ locally applied to the single level. The reservoir is an electron gas with temperature T and chemical potential μ , and the strength of the coupling between the two subsystems is arbitrary. The setup also includes a “floating contact”, which we will discuss in detail later on.

The effect of the periodic driving is twofold. On one hand, it induces a charge current that flows between the dot and the reservoir as a function of time. On the other hand, it performs a work on the system, thus injecting energy that is ultimately dissipated as heat deep inside the reservoir. Importantly, due to charge conservation, the electronic current is defined as the change in time of the electron number either at the reservoir or on the dot. No contribution of the tunneling region on the charge current exists. In contrast, the energy delivered by the external ac source is temporarily stored in three different parts of the setup: the dot, the reservoir and also in the dot-reservoir tunneling region. The role of the tunneling region is typically disregarded in classical thermodynamics because it is a surface term that is negligible when both the system and the reservoir are macroscopic [26]. Yet, in the nanoscale setup studied here the amount of energy stored in the dot is comparable to that of the tunneling region and the latter can no longer be neglected.

In a recent work [15] we have coined the name of *energy reactance* to characterize the energy temporarily stored at the tunneling region. This is a thermal analogue of an electrical reactance (due to electrical capacitance and inductance), which manifests itself in a time-dependent setup only. We have argued that it is physically meaningful to take the energy reactance into account as a contribution to the time-dependent heat current flowing into the reservoir. We have shown that this is in full agreement with the laws of thermodynamics [13, 23]. While some recent works raised some concerns [18, 22], other works arrived at conclusions similar to our analysis [14, 17, 21]. The aim of the present work is to make one step further by proposing a measurement scheme that is able to test the effect of the energy reactance onto a time-dependent heat flux.

Proposed experiment and predictions. The setup is sketched in Fig. 1, where we introduce a floating contact attached (e.g., via tunneling) to the quantum dot. When a periodic gate voltage $V(t)$ is applied, charge and heat currents enter not only the reservoir but also the floating contact. The latter can adjust its chemical potential μ_c and temperature T_c to maintain zero charge and heat currents flowing into it. We will focus on slow “adiabatic” driving, which corresponds to a driving period much larger than any characteristic time scale for the system. Assuming that the charge and energy relaxation rate of the floating contact is much faster than any other characteristic time, μ_c and T_c will change instantaneously to prevent charge and heat accumulation on the floating contact. In contrast, the reservoir is a mas-

sive electrode that keeps its temperature and chemical potential constant independently of the ac potential. In practice, this can be achieved grounding the reservoir as indicated in Fig. 1. Its temperature variations would be suppressed if the reservoir has in addition a large heat capacity.

The evolution of the chemical potential and temperature of the floating contact as the dot is adiabatically driven can be sensed by means of a voltage probe and a thermometer [27–35], as indicated in the figure. We predict different behaviors for μ_c and T_c depending on whether the energy reactance is considered or not in the heat flux into the floating contact. In this way, the proposed experiment would help to discern on the proper definition of the heat current and test the existence of the energy reactance.

The results are the following: (i) By defining the heat flux into the floating contact, taking into account the energy reactance as in Ref. 15, the temperature of the floating contact does not vary with time. The outcome is

$$T_c = T, \quad (1)$$

where T is the background temperature. The chemical potential of the contact $\mu_c(t)$ does vary with time in a periodic fashion with a period dictated by the electrical current flowing through the dot. (ii) We demonstrate that any other definition of the heat current, that does not properly account for the energy reactance, necessarily leads to a change in both quantities, $T_c(t)$ and $\mu_c(t)$ as functions of time.

Heat current into the floating contact and quantum energy reactance. Let the rates of change for the charge and the internal energy stored in the floating contact due to exchanges with the rest of the device be, respectively, $\dot{N}_c(t)$ and $\dot{U}_c(t)$. Similarly, the rate of change for the energy stored at the tunneling region between the dot and the floating contact is denoted by $\dot{U}_{T_c}(t)$. The meaningful definition for the instantaneous heat current entering the floating contact is [15]

$$\dot{Q}_c(t) = \dot{U}_c(t) + \frac{\dot{U}_{T_c}(t)}{2} - \mu_c(t)\dot{N}_c(t). \quad (2)$$

The energy reactance, $\dot{U}_{T_c}(t)/2$, contributes to the heat flux only instantaneously and as such vanishes when averaged over one driving period.

From the theoretical point of view, the energy reactance is necessary to derive an instantaneous Joule law for the heat current into a (single-channel) floating contact at low temperatures, $\dot{Q}_c(t) = R_q[\dot{N}_c(t)]^2$, with the universal charge relaxation resistance, the Büttiker resistance, $R_q = h/2e^2$ [1, 24]. The energy reactance is also necessary to both reconcile the relation between the Green function and the scattering matrix formalisms [25] for the instantaneous heat current [15] and also to obtain

correct frequency parity properties of the response functions [17].

Temperature and chemical potential of the floating contact. Our goal is to explicitly show that the definition of Eq. (2) can be verified by measuring the temperature and chemical potential of the floating contact. The latter is a small conductor that we assume to be in thermal equilibrium at every instant of time such that both temperature and chemical potential adjust themselves to satisfy the condition of vanishing charge and vanishing heat current, i.e., $\mu_c(t)$ and $T_c(t)$ simultaneously fulfill $\dot{Q}_c(t) = \dot{N}_c(t) = 0$. The local equilibrium condition is justified in the adiabatic regime (very low driving frequency Ω), mostly accessible in experiments [1]. Deviation of the floating contact temperature and chemical potential from their stationary values are denoted by $\delta T_c(t) = T_c(t) - T$ and $\delta\mu_c(t) = \mu_c(t) - \mu$, respectively. In the adiabatic regime, these quantities are small, $\delta T_c(t), \delta\mu_c(t) \propto \hbar\Omega$. As a consequence, we can evaluate both charge and heat fluxes in linear response in these quantities (while the amplitude of the ac driving potential is arbitrary).

Following Refs. 23 and 37 we expand the fluxes $\mathbf{J}(t) \equiv (\dot{N}_c(t), \dot{Q}_c(t))$ in the affinities $\mathbf{X} = (\delta\mu_c(t), \delta T_c(t), \hbar\Omega)$, with coefficients $\Lambda_{ij}(t)$ as

$$J_i(t) = \sum_{j=1}^3 \Lambda_{ij}(t) X_j(t), \quad (3)$$

where $i = 1, 2$ ($j = 1, 2, 3$) label the different components of the vectors \mathbf{J} and \mathbf{X} , respectively. The coefficients of the above expansion are response functions evaluated with the frozen Hamiltonian at time t and have the following physical interpretation: Λ_{11} and Λ_{22} are the usual electric and thermal conductances. On the other hand, Λ_{12} (related to the Seebeck effect) and Λ_{21} (related to the Peltier effect) capture the thermoelectric transport, and they satisfy the reciprocity relation $\Lambda_{21} = T\Lambda_{12}$ [38–41]. Finally, Λ_{13} and Λ_{23} describe, respectively, the generation of charge and heat currents by the ac driving. They also obey Onsager relations with the coefficients entering the work flux (not considered here) [37]. Explicit expressions of these coefficients will be supplied below for the specific model.

Here, we notice that the conditions of vanishing fluxes to the floating contact amounts to finding the solution of the 2×2 linear set of equations $\sum_{j=1}^2 \Lambda_{ij} X_j = -\Lambda_{i3} \hbar\Omega$, $i = 1, 2$. The solutions are

$$\begin{aligned} \delta\mu_c(t) &= \frac{\Lambda_{12}\Lambda_{23} - \Lambda_{13}\Lambda_{22}}{\det \mathbf{\Lambda}'} \hbar\Omega, \\ \delta T_c(t) &= \frac{\Lambda_{13}\Lambda_{21} - \Lambda_{11}\Lambda_{23}}{\det \mathbf{\Lambda}'} \hbar\Omega, \end{aligned} \quad (4)$$

where $\det \mathbf{\Lambda}'$ corresponds to the determinant of the 2×2 matrix determined by the condition $j \neq 3$.

The coefficients Λ can be calculated for the system considered in Fig. 1 following Refs. 25 and 36 (see details in Ref. 42)

$$\Lambda_{ij}(t) = \begin{cases} \int \frac{(\varepsilon - \mu)^{i+j-2}}{\hbar T^{(j-1)}} \mathcal{T}(t, \varepsilon) \partial_\varepsilon f d\varepsilon & \text{if } j \neq 3 \\ -\frac{\Gamma_c \dot{V}}{(\Gamma + \Gamma_c) \hbar \Omega} \int (\varepsilon - \mu)^{i-1} \rho_f(t, \varepsilon) \partial_\varepsilon f d\varepsilon & \text{if } j = 3, \end{cases} \quad (5)$$

The distinction between $j \neq 3$ and $j = 3$ is important. In the former case, the response depends on the instantaneous transmission probability $\mathcal{T}(t, \varepsilon)$ for electrons traversing the quantum dot between the reservoir and the floating contact. Physically, this corresponds to dc transport. In the latter case, the response is a function of the time derivative of the potential applied to the gate, $\dot{V} = -\Omega V \sin(\Omega t)$, and the instantaneous local density of states of the dot, $\rho_f(t, \varepsilon)$. Physically, this is pumping and, as such, of ac nature. Both coefficients are time dependent because the system adiabatically reacts to the instantaneous ac driving potential [43]. Finally, in Eq. (5) f is the Fermi-Dirac distribution of the reservoir, while $\Gamma_c = |w_c|^2 \rho_c$ and $\Gamma = |w|^2 \rho$ are the hybridization functions with w_c the dot-floating contact couplings and w the dot-reservoir couplings. The density of states of the floating contact is ρ_c and that of the reservoir is ρ .

Interestingly, we readily find that the coefficients of Eq. (5) satisfy *i)* $\Lambda_{13}\Lambda_{21} - \Lambda_{11}\Lambda_{23} = 0$ and *ii)* $\Lambda_{j3} = -\Lambda_{j1} \frac{\dot{V}}{\Gamma\Omega}$ with $j = 1, 2$, leading to the solution

$$\delta T_c(t) = 0, \quad \delta\mu_c(t) = \frac{\hbar}{\Gamma} e\dot{V}. \quad (6)$$

From Eq. (4) and the relation *ii)*, we see that the above results do not actually depend on coupling to the floating contact. We have checked that Eq. (6) is valid for any temperature T , provided that the adiabaticity condition $\Gamma, \Gamma_c \gg \hbar\Omega$ is satisfied [44, 45]. This is true even for temperatures close to zero, in which case the second order contributions in the affinities should be added to Eq. (3) [42].

In summary, the floating contact fulfills the conditions of vanishing heat and charge fluxes by changing $\delta\mu_c(t)$ in time according to Eq. (6) while keeping its temperature constant and equal to the background temperature, as indicated in Eq. (1).

Examine the energy reactance. We would like to stress now that Eq. (6), in particular, the prediction of a constant temperature of the floating contact expressed in Eq. (1), constitutes a proof for the existence of the energy reactance $\dot{U}_{T_c}(t)/2$ and the definition of the heat current as in Eq. (2). This can be easily understood by noticing that we would arrive at completely different conclusions on the behavior of the temperature of the floating contact if we consider a definition of the heat flux that does not take into account the energy reactance.

As a proof, let us analyze the consequence of adopting a commonly used definition, that does not take into

account the energy reactance. This corresponds to the following expression for the heat flux into the floating contact,

$$\dot{Q}_c(t) = \dot{U}_c(t) - \mu_c(t)\dot{N}_c(t). \quad (7)$$

We need to recalculate the coefficients $\Lambda_{2j}(t)$ by using the above equation. We denote the so defined coefficients by $\tilde{\Lambda}_{2,j}(t)$. From Eq. (4), where we replace $\Lambda_{2,j}(t) \rightarrow \tilde{\Lambda}_{2,j}(t)$, $j = 1, 2, 3$, we find the floating contact temperature $\delta\tilde{T}_c(t)$ and chemical potential $\delta\tilde{\mu}_c(t)$. In contrast to Eq. (6), now we find that both the temperature $\delta\tilde{T}_c(t)$ and the chemical potential $\delta\tilde{\mu}_c(t)$ of the floating contact change in time. In the case of the chemical potential, $\delta\tilde{\mu}_c$ evolves in time in a different pattern from that described by Eq. (6). We turn to focus on the behavior of the temperature $\delta\tilde{T}_c$, which is shown in Fig. 2. It is worth noting that the amplitude of the $\delta\tilde{T}_c$ oscillations decreases as T increases, which shows that the two definitions of the heat current agree in the high temperature limit. These results show that the role of the energy reactance is particularly relevant in the quantum regime. In the classical high-temperature limit the temperature of the floating contact is independent of time, either with the heat current defined as in Eq. (2) or with the definition of Eq. (7).

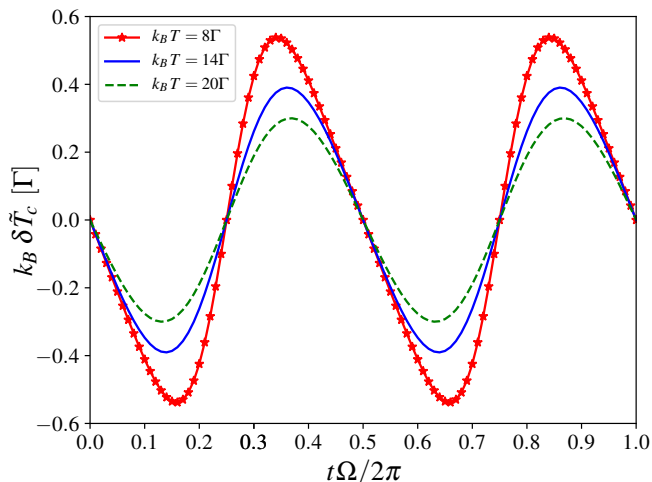


FIG. 2. Deviation of the temperature of the floating contact, $\delta\tilde{T}_c$, as a function of time for different background temperatures T . The ac potential is $20\Gamma\cos(\Omega t)$ with $\hbar\Omega = 0.07\Gamma$. The hybridization between the floating contact and the quantum level is $\Gamma_c = 0.6\Gamma$. All energies are expressed in units of the hybridization Γ with the reservoir. The temperature of the floating contact displays oscillations that depend on the background temperature. As T increases, the oscillations become less pronounced and the maxima positions deviate from the moment when the level is aligned with the chemical potential of the reservoir, which in this case corresponds to $t\Omega/2\pi = 0.25$ and $t\Omega/2\pi = 0.75$.

Conclusion. We have shown that the behavior of the time-resolved chemical potential and temperature of a floating contact coupled to an adiabatically driven quantum dot is strongly sensitive on the definition of the instantaneous heat flux. For this reason, sensing these quantities would provide an experimental test for the relevance of the energy reactance introduced in Ref. [15] as a component of the time-dependent heat flux.

Specifically, for an adiabatically driven quantum dot with a single active level coupled to a single reservoir, we have shown that: (i) If the energy reactance is taken into account, then the temperature of the floating contact is constant and equal to that of the reservoir, while its chemical potential follows the time derivative of the driving potential, \dot{V} , as expressed in Eq. (6). Instead, (ii) if the energy reactance is not taken into account, these two quantities follow a nonuniversal and rather cumbersome time-dependent pattern.

The experiment we propose is close to the scope of present-day experimental techniques. In fact, typical level spacing for quantum dots is around $100\mu\text{eV}$ [46]. Thus, by keeping driving amplitudes below this energy, we would basically have a single active level. On the other hand, typical parameters for single particle emitters have $\Gamma \simeq 1\mu\text{eV}$ ($\simeq 1\text{GHz}$) and are operated at frequencies $\Omega \simeq 0.1\text{GHz}$ [1], which satisfy the adiabatic condition $\hbar\Omega < \Gamma$. As a consequence, a fast thermometer [9] is able to follow temperature changes of the floating contact on the nanosecond scale. Experiments are typically performed at temperatures close to $T \sim 100\text{mK}$. For this temperature, the oscillations in the temperature shown in Fig. 2 have an amplitude of $\delta\tilde{T}_c \simeq 10\text{mK}$.

We emphasize that the question about the role of the energy reactance in the definition of a time-dependent heat flux is a fundamental one. It is not restricted to slowly driven systems of noninteracting electrons but is also relevant for interacting models, for fast drivings, and for weakly and strongly coupled systems. So far this question has been addressed only theoretically. The present proposal shows that a thermometer probe response will experimentally demonstrate the existence of the energy reactance.

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- [1] J. Gabelli, G. Fève, J.-M. Berroir, B. Plaças, A. Cavanna, B. Etienne, Y. Jin, and D. C. Glattli, Violation of Kirchhoff's laws for a coherent RC circuit, *Science* **313**, 499–502 (2006).
 [2] G. Fève, A. Mahé, J.-M. Berroir, T. Kontos, B. Plaças,

- D. C. Glattli, A. Cavanna, B. Etienne, and Y. Jin, An on-demand coherent single-electron source, *Science* **316**, 1169–1172 (2007).
- [3] J. Dubois, T. Jullien, F. Portier, P. Roche, A. Cavanna, Y. Jin, W. Wegscheider, P. Roulleau, and D. C. Glattli, Minimal-excitation states for electron quantum optics using levitons. *Nature*, **502**, 659–663 (2013).
- [4] S. Hermelin, S. Takada, M. Yamamoto, S. Tarucha, A. D. Wieck, L. Saminadayar, C. Bäuerle, and T. Meunier, Electrons surfing on a sound wave as a platform for quantum optics with flying electrons, *Nature* **477**, 435–438 (2011).
- [5] R. P. G. McNeil, M. Kataoka, C. J. B. Ford, C. H. W. Barnes, D. Anderson, G. A. C. Jones, I. Farrer, and D. A. Ritchie, On-demand single-electron transfer between distant quantum dots, *Nature* **477**, 439–442 (2011).
- [6] B. Bertrand, S. Hermelin, P.-A. Mortemousque, S. Takada, M. Yamamoto, S. Tarucha, A. Ludwig, A. D. Wieck, C. Bäuerle, and T. Meunier, Injection of a single electron from static to moving quantum dots, *Nanotechnology* **27**, 214001 (2016).
- [7] J. D. Fletcher, P. See, H. Howe, M. Pepper, S. P. Giblin, J. P. Griffiths, G. A. C. Jones, I. Farrer, D. A. Ritchie, T. J. B. M. Janssen, and M. Kataoka, Clock-Controlled Emission of Single-Electron Wave Packets in a Solid-State Circuit, *Physical Review Letters* **111**, 216807 (2013).
- [8] For the most recent advancements of the field see the special issue: J. Splettstoesser and R. J. Haug, Single-electron control in solid state devices, *Phys. Stat. Sol. (b)* **254**, 1770217–2 (2017).
- [9] M. Zgirski, M. Foltyn, A. Savin, M. Meschke, and J. P. Pekola, Nanosecond thermometry with Josephson junction, arXiv:1704.04762.
- [10] J. P. Pekola, P. Solinas, A. Shnirman, and D. V. Averin, Calorimetric measurement of work in a quantum system, *New J. Phys.* **15**, 115006 (2013).
- [11] S. Gasparinetti, K. L. Viisanen, O.-P. Saira, T. Faivre, M. Arzeo, M. Meschke, and J. P. Pekola, Fast Electron Thermometry for Ultrasensitive Calorimetric Detection, *Physical Review Applied* **3**, 014007 (2015).
- [12] R. Kosloff, Quantum Thermodynamics: A Dynamical Viewpoint, *Entropy* **15**, 2100–2128 (2013).
- [13] M. Ludovico, L. Arrachea, M. Moskalets, and D. Sánchez, Periodic Energy Transport and Entropy Production in Quantum Electronics, *Entropy* **18**, 419 (2016).
- [14] L.-A. Wu and D. Segal, Energy flux operator, current conservation and the formal Fourier’s law, *J. Phys. A: Math. Theor* **42**, 025302 (2008).
- [15] M. F. Ludovico, J.S. Lim, M. Moskalets, L. Arrachea, and D. Sánchez, Dynamical energy transfer in ac-driven quantum systems, *Phys. Rev. B* **89**, 161306 (2014).
- [16] J. Ankerhold and J. P. Pekola, Heat due to system-reservoir correlations in thermal equilibrium, *Physical Review B* **90**, 075421 (2014).
- [17] G. Rosselló, R. López, and J. S. Lim, Time-dependent heat flow in interacting quantum conductors, *Phys. Rev. B* **92**, 115402 (2015).
- [18] M. Esposito, M. A. Ochoa, and M. Galperin, Nature of heat in strongly coupled open quantum systems, *Physical Review B* **92**, 235440 (2015).
- [19] M. Esposito, M. A. Ochoa, and M. Galperin, Quantum Thermodynamics: A Nonequilibrium Green’s Function Approach, *Physical Review Letters* **114**, 080602 (2015).
- [20] A. M. Daré and P. Lombardo, Time-dependent thermoelectric transport for nanoscale thermal machines, *Physical Review B* **93**, 035303 (2016).
- [21] A. Bruch, M. Thomas, S. V. Kusminskiy, F. von Oppen, and A. Nitzan, Quantum thermodynamics of the driven resonant level model, *Physical Review B* **93**, 115318 (2016).
- [22] M. A. Ochoa, A. Bruch, and A. Nitzan, Energy distribution and local fluctuations in strongly coupled open quantum systems: The extended resonant level model, *Physical Review B* **94**, 035420 (2016).
- [23] M. F. Ludovico, M. Moskalets, D. Sánchez, and L. Arrachea, Dynamics of energy transport and entropy production in ac-driven quantum electron systems, *Phys. Rev. B* **94**, 035436 (2016).
- [24] M. Büttiker, H. Thomas, A. Prêtre, Mesoscopic capacitors, *Physics Letters A* **180**, 364–369 (1993).
- [25] L. Arrachea, Exact Green’s function renormalization approach to spectral properties of open quantum systems driven by harmonically time-dependent fields, *Phys. Rev. B* **75**, 035319 (2007).
- [26] M. Le Bellac, F. Montessagne, G. G. Batrouni, *Equilibrium and Non-Equilibrium Statistical Thermodynamics* (Cambridge University Press, Cambridge, 2004).
- [27] H. L. Engquist and P. Anderson, Definition and measurement of the electrical and thermal resistance, *Physical Review B* **24**, 1151 (1981).
- [28] Y. Dubi and M. Di Ventra, Thermoelectric Effects in Nanoscale Junctions, *Nano Lett.* **9**, 97 (2009).
- [29] K. Saito, G. Benenti, G. Casati, and T. Prosen, Thermopower with broken time-reversal symmetry, *Phys. Rev. B* **84**, 201306(R) (2011).
- [30] D. Sánchez and L. Serra, Thermoelectric transport of mesoscopic conductors coupled to voltage and thermal probes, *Phys. Rev. B* **84**, 201307(R) (2011).
- [31] J. P. Bergfield, S. M. Story, R. C. Stafford, and C. A. Stafford, Probing Maxwell’s Demon with a Nanoscale Thermometer, *ACS Nano* **7**, 4429 (2013).
- [32] F. Battista, M. Moskalets, M. Albert, and P. Samuelsson, Quantum Heat Fluctuations of Single-Particle Sources, *Physical Review Letters* **110**, 126602 (2013).
- [33] A. Caso, L. Arrachea, and G. S. Lozano, Local temperatures and heat flow in quantum driven systems, *Phys. Rev. B* **83**, 165419 (2011).
- [34] L. Arrachea and E. Fradkin, Chiral heat transport in driven quantum Hall and quantum spin Hall edge states, *Phys. Rev. B* **84**, 235436 (2011).
- [35] L. Ye, D. Hou, X. Zheng, Y. Yan, and M. Di Ventra, Local temperatures of strongly-correlated quantum dots out of equilibrium, *Phys. Rev. B* **91**, 205106 (2015).
- [36] L. Arrachea and M. Moskalets, Relation between scattering matrix and Keldysh formalisms for quantum transport driven by time-periodic fields, *Phys. Rev. B* **74**, 245322 (2006).
- [37] M. F. Ludovico, F. Battista, F. von Oppen, and L. Arrachea, Adiabatic response and quantum thermoelectrics for ac-driven quantum systems, *Physical Review B* **93**, 075136 (2016).
- [38] L. Onsager, Reciprocal relations in irreversible processes, *Phys. Rev.* **37**, 404 (1931).
- [39] M. Büttiker, Four-Terminal Phase-Coherent Conductance, *Phys. Rev. Lett.* **57**, 1761 (1986).
- [40] J. Matthews, F. Battista, D. Sánchez, P. Samuelsson, and H. Linke, Experimental verification of reciprocity relations in quantum thermoelectric transport, *Phys. Rev. B* **90**, 165428 (2014).

- [41] G. Benenti, G. Casati, K. Saito, and R. Whitney, Fundamental aspects of steady-state conversion of heat to work at the nanoscale, arXiv:1608.05595.
- [42] See Supplementary Material for details of the parameters entering the model, the calculation of the charge and heat currents, and explicit expressions of the coefficients in the linear and second order expansions.
- [43] M.V. Moskalets, *Scattering Matrix Approach to Non-Stationary Quantum Transport*. (World Scientific, London, 2012).
- [44] P. W. Brouwer, Scattering approach to parametric pumping Phys. Rev. B **58**, 10135 (R) (1998).
- [45] J. Splettstoesser, S. Ol'khovskaya, M. Moskalets, and M. Büttiker, Electron counting with a two-particle emitter, Phys. Rev. B **78**, 205110 (2008).
- [46] L. P. Kouwenhoven, C. M. Marcus, P. L. McEuen, S. Tarucha, R. M. Westervelt, N. S. Wingreen, Electron transport in quantum dots, in *Mesoscopic Electron Transport*, ed. by L. L. Sohn, L. P. Kouwenhoven and G. Schön (Kluwer, Dordrecht, 1996).

**PROBING THE ENERGY REACTANCE WITH ADIABATICALLY DRIVEN QUANTUM DOTS:
SUPPLEMENTARY INFORMATION**

This supporting document describes in detail the derivation of the time-dependent linear response coefficients for particle and heat currents flowing into the floating contact.

Theoretical model

We consider a simple setup, with all the necessary ingredients to analyze the dynamical energy transfer and to probe the energy reactance. It is the most basic model for a quantum capacitor, which consists in a periodically driven single level (quantum dot) coupled to an electron bath, the reservoir, with temperature $T_r = T$ and chemical potential $\mu_r = \mu$. The time-dependent driving is provided by the application of an oscillatory gate voltage of the form $V(t) = V\cos(\Omega t)$.

To test the effect of the energy reactance on the time-dependent heat flux, we introduce a floating contact, which is coupled to the driven level. In order to be electrically and thermally isolated from the environment at every time, the floating contact instantaneously adjusts its chemical potential μ_c and temperature T_c .

The Hamiltonian of the full system, the quantum capacitor together with the floating contact, can be separated into three contributions,

$$H(t) = H_{QC}(t) + H_c + H_{T_c}. \quad (S1)$$

The Hamiltonian H_{QC} represents the quantum capacitor, which contains three elements: the single level, an electron bath (the reservoir, denoted by the letter r), and the coupling between the two. Then,

$$H_{QC}(t) = \sum_k \left[\varepsilon_k^r c_k^\dagger c_k + w(d^\dagger c_k + c_k^\dagger d) \right] + (\varepsilon_0 + V(t))d^\dagger d, \quad (S2)$$

where ε_k^r is the energy band of the reservoir and w is the coupling amplitude to the driven level. The energy ε_0 corresponds to the bare level, which for simplicity will be considered aligned with the chemical potential of the reservoir, i.e. $\varepsilon_0 = \mu$. The operator c_k^\dagger (c_k) create (destroy) an electron with a wavevector k in the reservoir, while d^\dagger and d are associated to the degrees of freedom of the single level.

Similarly, the floating contact is represented by the Hamiltonian,

$$H_c = \sum_q \varepsilon_q^c a_q^\dagger a_q, \quad (S3)$$

while in this case, the operators a_q and a_q^\dagger are responsible, respectively, for the creation and destruction of an electron in the floating contact with an energy ε_q^c . The coupling between the level and the floating contact can be written as

$$H_{T_c} = \sum_q w^c (d^\dagger a_q + a_q^\dagger d). \quad (S4)$$

Particle and heat currents

Now, the aim is to compute the particle and heat fluxes entering the floating contact. The time variation of the particles present in the floating contact is given by the exact expression, $\dot{N}_c = \frac{i}{\hbar} \langle [H, N_c] \rangle$. To compute the time-dependent heat current as in Eq. (2), or by adopting the definition (7), we define the internal energy stored in the floating contact and in the tunneling region as $\dot{U}_\beta = \frac{i}{\hbar} \langle [H, H_\beta] \rangle$, with $\beta = c, T_c$.

In Refs. [1] and [2], we presented that the evolution of the expectation value of an observable (e.g the number of particles or the energies) can be obtained by recourse to Keldysh non-equilibrium Green's functions. In this way, the different currents can be computed in terms of the retarded Green function $G^R(t, t') = -i\theta(t - t') \langle \{d(t), d^\dagger(t')\} \rangle$ of the single level. For a time-periodic driving it is convenient to use the Floquet-Fourier representation [3],

$$G^R(t, t') = \sum_{n=-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{d\varepsilon}{2\pi} e^{-i\frac{\varepsilon}{\hbar}(t-t')} e^{-in\Omega t} \mathcal{G}(n, \varepsilon). \quad (S5)$$

As we showed in detail in Ref. [1] for a single driven level connected to many reservoirs, the particle current entering one of them, as for example the floating contact, is

$$\dot{N}_c(t) = \sum_l \int \frac{d\varepsilon}{h} e^{-i\Omega t} \Gamma_c \left\{ i\mathcal{G}^*(-l, \varepsilon) [f_c(\varepsilon) - f_c(\varepsilon_l)] - \sum_n \sum_{\alpha=r,c} [f_c(\varepsilon) - f_\alpha(\varepsilon_n)] \Gamma_\alpha \mathcal{G}(l+n, \varepsilon_n) \mathcal{G}^*(n, \varepsilon_n) \right\}, \quad (\text{S6})$$

where $\alpha = r$ corresponds to the reservoir of the capacitor, and $\alpha = c$ is the floating contact. Some of the energies are shifted by a multiple of the energy quantum $\hbar\Omega$ as $\varepsilon_n = \varepsilon - n\hbar\Omega$. We have introduced the Fermi-Dirac distribution of the reservoir labeled by α , $f_\alpha(\varepsilon) = [e^{(\varepsilon - \mu_\alpha)/k_B T_\alpha} + 1]^{-1}$, with k_B being the Boltzmann constant. The hybridizations with the reservoirs are $\Gamma_c = |w^c|^2 \rho_c$ for the floating contact and $\Gamma = |w|^2 \rho$ for the reservoir, with $\rho_c = \sum_{k \in c} 2\pi \delta(\varepsilon - \varepsilon_k^c)$ and $\rho = \sum_{k \in m} 2\pi \delta(\varepsilon - \varepsilon_k^m)$ being the density of states of the floating contact and the reservoir, respectively. We are considering the wide band limit, in which Γ_c and Γ are constant functions.

In the same work [1], and also in Ref. [2] for a more general setup, we showed that according to the definition in Eq. (2) the heat flux is

$$\begin{aligned} \dot{Q}_c(t) = \sum_l \int \frac{d\varepsilon}{h} e^{-i\Omega t} \Gamma_c \left\{ i\mathcal{G}^*(-l, \varepsilon) (\varepsilon_{\frac{l}{2}} - \mu_c) [f_c(\varepsilon) - f_c(\varepsilon_l)] \right. \\ \left. - \sum_n \sum_{\alpha=r,c} (\varepsilon_{-\frac{l}{2}} - \mu_c) [f_c(\varepsilon) - f_\alpha(\varepsilon_n)] \Gamma_\alpha \mathcal{G}(l+n, \varepsilon_n) \mathcal{G}^*(n, \varepsilon_n) \right\}. \end{aligned} \quad (\text{S7})$$

If we adopt the definition in Eq. (7), which does not take into account the energy reactance $\dot{U}_{T_c}(t)/2$, then

$$\begin{aligned} \dot{Q}_c(t) = \sum_l \int \frac{d\varepsilon}{h} e^{-i\Omega t} \Gamma_c \left\{ i\mathcal{G}^*(-l, \varepsilon) \left[(\varepsilon - \mu_c) (f_c(\varepsilon) - f_c(\varepsilon_l)) + l\hbar\Omega f_c(\varepsilon_l) \right] \right. \\ \left. - \sum_n \sum_{\alpha=r,c} \left[(\varepsilon - \mu_c) (f_c(\varepsilon) - f_\alpha(\varepsilon_n)) - \frac{l}{2} \hbar\Omega f_\alpha(\varepsilon_n) \right] \Gamma_\alpha \mathcal{G}(l+n, \varepsilon_n) \mathcal{G}^*(n, \varepsilon_n) \right\}. \end{aligned} \quad (\text{S8})$$

Here, we stress that all the above expressions for both particle and heat fluxes are exact, in the sense that they are valid for arbitrary values of the driving frequency, amplitude and temperature.

Linear response coefficients Λ and $\tilde{\Lambda}$

In what follows, we focus on the adiabatic regime, in which the driving frequency Ω is very low. As we presented in our previous works [1, 2], to which we refer the reader for further details, we can expand the Floquet components $\mathcal{G}(n, \varepsilon)$ up to first order in Ω as

$$\mathcal{G}(n, \varepsilon) \sim \int_0^\tau \frac{dt}{\tau} e^{in\Omega t} \left[G^f(t, \varepsilon) + \frac{i\hbar}{2} \frac{\partial^2}{\partial t \partial \varepsilon} G^f(t, \varepsilon) \right]. \quad (\text{S9})$$

Here $\tau = 2\pi/\Omega$ is the driving period, and

$$G^f(t, \varepsilon) = \left(\varepsilon - \varepsilon_0 - V(t) - i \frac{(\Gamma + \Gamma_c)}{2} \right)^{-1} \quad (\text{S10})$$

is the frozen Green's function, which corresponds to the equilibrium solution of the Dyson equation [4] at a given *frozen* time t .

Within the adiabatic regime, the departures of the temperature and chemical potential of the floating contact from those of the reservoir, δT_c and $\delta \mu_c$, are proportional to $\hbar\Omega$. Hence, we can also evaluate Eqs. (S6), (S7) and (S8) within linear response in these quantities by expanding

$$f_c(\varepsilon_n) \sim f(\varepsilon) - \partial_\varepsilon f n\hbar\Omega - \partial_\varepsilon f \delta \mu_c - \frac{(\varepsilon - \mu)}{T} \partial_\varepsilon f \delta T_c \quad (\text{S11})$$

where $f_r(\varepsilon) = f(\varepsilon)$ is the Fermi distribution of the reservoir.

Then, by using the expansions for slow driving (S9) and (S11) in the expressions of the charge and heat fluxes, we can compute the linear response coefficients Λ as defined in Eq. (3) of the main text

$$\Lambda_{ij}(t) = \begin{cases} \int \frac{(\varepsilon - \mu)^{i+j-2}}{hT^{(j-1)}} \mathcal{T}(t, \varepsilon) \partial_\varepsilon f d\varepsilon & \text{if } j \neq 3 \\ -\frac{\Gamma_c \dot{V}}{(\Gamma + \Gamma_c)h\Omega} \int (\varepsilon - \mu)^{i-1} \rho_f(t, \varepsilon) \partial_\varepsilon f d\varepsilon & \text{if } j = 3, \end{cases} \quad (5)$$

where $\rho_f(t, \varepsilon) = |G^f(t, \varepsilon)|^2 (\Gamma + \Gamma_c)$ is the total frozen density of states of the quantum dot, and $\mathcal{T}(t, \varepsilon) = |G^f(t, \varepsilon)|^2 \Gamma_c \Gamma$ is the transmission probability.

The same procedure can be applied to the heat flux in Eq. (S8), whose definition does not take into account the energy stored in the tunneling region. In this case, we replace $\Lambda^{2j} \rightarrow \tilde{\Lambda}^{2j}$, so that

$$\dot{Q}_c(t) = \sum_{j=1}^3 \tilde{\Lambda}_{2j}(t) X_j(t). \quad (S12)$$

For this different definition of the heat, we find that $\tilde{\Lambda}_{21}(t) = \Lambda_{21}(t)$ and $\tilde{\Lambda}_{22}(t) = \Lambda_{22}(t)$, while the coefficient describing the pumping of heat changes as

$$\tilde{\Lambda}_{23}(t) = -\frac{\Gamma_c \dot{V}}{(\Gamma + \Gamma_c)h\Omega} \int d\varepsilon \frac{df}{d\varepsilon} \rho_f(t, \varepsilon). \quad (S13)$$

Chemical potential and temperature of the floating contact in the zero temperature limit

In the very low temperature limit of the reservoir, when $T \rightarrow 0$, an analysis of the fluxes within linear response turns out not to be appropriate anymore since second order contributions could become dominant. In this case, instead of Eq. (3), the fluxes should be expanded as

$$J_i = \sum_{j=1}^3 \left(\Lambda_{ij} X_j + \sum_{m \leq j} L_{mj}^i X_m X_j \right), \quad (S14)$$

where \vec{L} is a vector composed by matrices which capture the second order terms. The extreme situation occurs at $T = 0$, in which absolutely all the linear response coefficients of the heat \dot{Q}_c vanish ($\Lambda_{2j} = 0$). In what follows, we focus on that case and compute both heat and particle fluxes entering the floating contact, in order to study if Eq. (6) remain valid when the temperature of the reservoir is close to zero. For the heat flux we find

$$\dot{Q}_c^{T=0}(t) = L_{22}^2(t) \delta T_c(t)^2 + L_{13}^2(t) \delta \mu_c(t) \hbar \Omega + L_{33}^2(t) (\hbar \Omega)^2, \quad (S15)$$

where

$$\begin{aligned} L_{22}^2(t) &= -\frac{\pi^2}{3h} \mathcal{T}(t, \mu) \\ L_{13}^2(t) &= -\frac{\Gamma_c \Gamma \dot{V}}{(\Gamma + \Gamma_c)h\Omega} \frac{\rho_f(t, \mu)^2}{2}, \\ L_{33}^2(t) &= -\frac{\dot{V}}{\Gamma \Omega} L_{13}^2(t) \end{aligned} \quad (S16)$$

and all other coefficients are zero, $L_{11}^2 = L_{12}^2 = L_{23}^2 = 0$. However for the particle current, unlike the heat flux, only the coefficient $\Lambda_{12} = 0$ at zero temperature while first order contributions in $\delta \mu_c$ and $\hbar \Omega$ remain. Thus, we can express the particle current at lowest order in the affinities as

$$\dot{N}_c^{T=0}(t) = \Lambda_{11}(t) \delta \mu_c(t) + \Lambda_{13}(t) \hbar \Omega + L_{22}^1(t) \delta T_c(t)^2, \quad (S17)$$

with

$$L_{22}^1(t) = -\frac{\pi^2}{3h} \partial_\varepsilon \mathcal{T}(t, \mu). \quad (S18)$$

Here, it is worth to mention that $L_{12}^1 = L_{23}^1 = 0$, so that $L_{22}^1 \delta T_c^2$ is the only lowest order contribution in δT_c .

Similarly, the chemical potential and the temperature of the floating contact can also be found by the condition $\dot{N}_c^{T=0} = \dot{Q}_c^{T=0} = 0$. In this occasion, the vanishing fluxes condition leads to a set of equation which is linear en δT_c^2 and $\delta \mu_c$, with solution

$$\delta T_c(t)^2 = \frac{L_{13}^2 \Lambda_{13} - \Lambda_{11} L_{33}^2}{(\Lambda_{11} L_{22}^2 - L_{22}^1 L_{13}^2 \hbar \Omega)} (\hbar \Omega)^2,$$

$$\delta \mu_c(t) = \frac{L_{22}^1 L_{33}^2 (\hbar \Omega)^2 - L_{22}^2 \Lambda_{13} \hbar \Omega}{(\Lambda_{11} L_{22}^2 - L_{22}^1 L_{13}^2 \hbar \Omega)}. \quad (\text{S19})$$

From Eqs. (S16) and (5), it is easy to notice that $L_{13}^2 \Lambda_{13} - \Lambda_{11} L_{33}^2 = 0$, and then

$$\delta T_c(t) = 0. \quad (\text{S20})$$

Moreover, the relation between L_{13}^2 and L_{33}^2 in Eq. (S16) and $\Lambda_{13} = -\frac{\dot{V}}{\Gamma \Omega} \Lambda_{11}$ in (5), lead to the solution

$$\delta \mu_c(t) = \frac{\hbar}{\Gamma} \dot{V} + \mathcal{O}(\Omega^2). \quad (\text{S21})$$

Therefore, we find that Eq. (6) obtained for $k_B T \gg \hbar \Omega$ within linear response regime, remain valid at zero temperature for which higher order contributions take place. This is a strong result, that shows the universality of the behavior of δT_c and $\delta \mu_c$ for any temperature of the reservoir.

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- [1] M. F. Ludovico, M. Moskalets, D. Sánchez, and L. Arrachea, Dynamics of energy transport and entropy production in ac-driven quantum electron systems, *Phys. Rev. B* **94**, 035436 (2016).
 - [2] M. Ludovico, L. Arrachea, M. Moskalets, and D. Sánchez, Periodic Energy Transport and Entropy Production in Quantum Electronics, *Entropy* **18**, 419 (2016).
 - [3] L. Arrachea, Exact Green's function renormalization approach to spectral properties of open quantum systems driven by harmonically time-dependent fields, *Phys. Rev. B* **75**, 035319 (2007).
 - [4] L. Arrachea and M. Moskalets, Relation between scattering matrix and Keldysh formalisms for quantum transport driven by time-periodic fields, *Phys. Rev. B* **74**, 245322 (2006).
 - [5] M. F. Ludovico, J.S. Lim, M. Moskalets, L. Arrachea, and D. Sánchez, Dynamical energy transfer in ac-driven quantum systems, *Phys. Rev. B* **89**, 161306 (2014).