Charge fluctuations in nonlinear heat transport

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4JARA—Fundamentals of Future Information Technology

(Dated: August 1, 2014)

We show that charge fluctuation processes are crucial for the nonlinear heat conductance through an interacting nanostructure, even far from a resonance. The often made assumption that off-resonant transport proceeds only by virtual occupation of charge states, underlying exchange-scattering models of transport, can fail dramatically for heat transport as compared to charge transport. This indicates that nonlinear heat transport spectroscopy may be a very promising experimental tool, in particular when combining energy-level control with recent advances in nanoscale thermometry that allow accurate measurements of heat currents. It provides new qualitative information about relaxation processes which can go unnoticed by the traditional charge-current measurements, for instance by strong negative differential heat conductance at positive heat current.

PACS numbers: 73.23.Hk, 73.63.-b, 73.50.Lw

Driven by the technologically important issue of dissipation of heat, the understanding of thermoelectric processes on the nanoscale is currently of great interest.1–4 Even for a microscopic device the electric current response to a thermal bias can be measured. This way, information like the type of carriers, i.e., holes vs. electrons, dominating the transport can be obtained.5 How- ever, this is not enough to understand the processes limiting the thermoelectric efficiency and the heat current must be considered.6–9 Even in the absence of a thermal bias the dependence of the heat current on the voltage bias and on the energy levels of the nanostructure is interesting but is experimentally more challenging to access. A recent breakthrough10 has allowed for measurements that are accurate enough to determine the asymmetry of heat dissipation and its relation to the electronic transmission characteristics, even for molecular junctions with conductances as low as $10^{-3}$ in units of $e^2/h$. Although experimentally even more challenging, additionally integrating energy-level control into thermoelectric junctions, e.g., by electrical gating, does not seem out of reach and is—as we will argue—very promising.

Gate controlled charge-current spectroscopy is a well-developed experimental tool to access the discrete quantum levels of nanostructures. There are two prominent features in the charge current driven by a source-drain voltage that underpin this successful method. The first depends on the level position relative to the electrochemical potentials: The current shows sharp changes as new resonant transport processes are switched on with increasing bias. These processes—are often referred to as single-electron tunneling (SET)—are readily identified in a three-terminal setup by plotting the charge conductance as function of the applied bias $V$ and the gate voltage, as exemplified in Fig. 1(a). Two-terminal measurements correspond to line traces through such a plot. The second type of resonance is independent of the level position and appears as a horizontal line at $V = \Delta$ since it originates in the inelastic excitation by an energy $\Delta$ at fixed local electron number. This off-resonant feature requires a next-order tunnel process in which other charge states are only visited virtually, known as inelastic electron tunneling (IETS)10,11 or inelastic cotunneling (ICOT)12,13. Under appropriate conditions this allows for a description in terms of effective exchange- and potential-scattering amplitudes. As $\Delta$ goes to zero, a nonequilibrium Kondo resonance develops at low temperature due to the renormalization of these amplitudes.14–16 Although $\Delta$ also appears in additional resonant tunneling features, its inelastic signature has the advantage of being much sharper at low temperatures due to its higher-order origin.11,12 This inelastic excitation provides access to a range of physical phenomena: the energy $\Delta$ can be an electronic level splitting (e.g., in a semiconductor nanostructure), carbon nanotube,17,18 or a dopant atom;19 a quantized vibrational frequency,20 a spin-splitting due to a magnetic field21,22, or exchange interaction23,24 or magnetic anisotropy (e.g., in nanomagnets),25,26 or ad-atoms,27,28, or spin-orbit coupling.29

Thermoelectric transport has also been investigated within the two above mentioned physical pictures. Theoretical works have mostly focused on the thermopower in the linear-response regime. For quantum dots, this includes the study of resonant tunneling30 and inelastic tunneling31,32, Kondo processes.33 Previous works addressing the nonlinear regime have either applied effective single-particle description34,35 or focused on thermoelectric devices close to resonance assuming weak tunneling36 or weak interaction.37 The heat current received much less attention.38 A classification of nonlinear heat transport features for a strongly interacting nanostructure going beyond weak tunneling, matching that of charge transport39,40 still seems to be missing. This is of importance not only for thermoelectric setups.
FIG. 1. (Color online) (a) Charge conductance $\partial I_C/\partial V$ and (b) energy conductance $\partial I_E/\partial V$ of a quantum dot with $U = 330 T$, $\Delta = U/4 = 83 T$, and $\Gamma = 3.3 \cdot 10^{-3} T$. Labels (i)–(vii) indicate the features discussed in the text which also appear at horizontally mirrored positions (not labeled). To deal with the sign change of $\partial I_E/\partial V$ we plot $l((\partial I_E/\partial V))$ where $l(x) = \text{sgn}(x) \log_{10}|x|$ for $|x| > 10$ and $l(x) = ax/10$ for $|x| < 10$ with $a = 2 \cdot 10^9$. Inset to (b): Linear energy conductance versus $\varepsilon + U/2$ around the right SET resonance.

with gate voltage control, but also for scanning probe setup\cite{44} in which the level position plays a crucial role.

In this Letter we address this problem and show that the heat current driven by a nonlinear electric and/or thermal bias contains new qualitative information and deviates in a striking way from the charge transport, both it sign and amplitude. Its dependence on the level position reveals that certain relaxation processes, commonly thought to be relevant only at resonance, can be very important for heat transport far from resonance by competing with an inelastic excitation at finite voltage bias $V = \Delta$. The onset of this competition is not determined by the broadening of the resonances (thermal- or tunnel-induced) but instead set by the inelastic excitation energy $\Delta$. Even far from a resonance, real occupation of more than one charge state is both possible and important. We illustrate this for a resonant level with strong Coulomb interaction and a well-defined spin-flip excitation $\Delta$ due to an external field. This suffices to classify nonlinear thermoelectric transport features that should generally be observable in a range of nanostructures with more complex spectra.

Energy transport.—To obtain a generic picture of thermoelectric transport through an interacting nanoscale object—for short a quantum dot—we analyze the nonlinear thermoelectric transport properties of the Anderson model $H = H_A + H_{\text{res}} + H_C$. The dot is described by $H_A = \sum_\sigma (\varepsilon + \sigma \Delta/2) d_\sigma \dagger d_\sigma + U n(n-1)/2$, where $d_\sigma$ with $\sigma = \uparrow, \downarrow$ are the electron operators on the dot. Here $\varepsilon = (\varepsilon_\uparrow + \varepsilon_\downarrow)/2$ is the orbital energy level and $\Delta = \varepsilon_\uparrow - \varepsilon_\downarrow$ denotes the energy of a local spin excitation due to a magnetic field for fixed $n = 1$ with electron number $n = \sum_\sigma d_\sigma \dagger d_\sigma$. Furthermore, $U$ is the strong Coulomb interaction penalty paid when counting $n = 2$. The electrodes, indexed by $\alpha = \text{L,R}$, are described as noninteracting reservoirs, $H_{\text{res}} = \sum_\sigma H_\sigma = \sum_\sigma c_\sigma \dagger c_\sigma$, with electron operators $c_\sigma$. We allow for both a nonlinear voltage bias $V$ between the reservoirs through their electrochemical potentials $\mu_{\text{L,R}} = \pm V/2$ as well as a nonlinear thermal bias through their temperatures $T_{\text{L,R}}$. The tunnel coupling has the generic form $H_t = t \sum_{k\sigma} (c_\sigma \dagger d_\sigma + \text{h.c.})$, the bare resonance width is given by $\Gamma = 2\pi\nu_0$, with $\nu_0$ the density of states in the reservoirs, and we set $e = h = k_B = g\mu_B = 1$. The charge current is $I_C = \langle \sum c_\sigma^{\dagger} c_\sigma \rangle$, where $N_R$ is the electron number operator of the reservoir $\alpha = \text{R}$. Similarly, the current is defined via $I_E = \langle \sum \sigma \dagger \upsilon_\sigma \rangle$. The measurable heat current can be obtained via $I_Q = I_E - \mu_B I_C$. We will only consider the stationary currents entering the right reservoir. Since experimentally the way the voltage is applied is known (here $\mu_B = -V/2 = -\mu_L$), the conserved charge current is available, the conversion from $I_Q$ to $I_E$ amounts to a simple background subtraction. We focus on the contribution $I_E$ since we find that it contains all interesting physical features which can dominate over those due to the charge current, see e.g., Fig. 2(c) below. Also, $I_C$ and $I_E$ are more easily compared, highlighting the differences in charge and heat transport most directly, in particular the bias and gate voltage dependence on which we focus here. The stationary currents and the underlying state occupations are calculated using a reduced density operator approach\cite{25, 26} accounting for the strong local interaction $U$. We go beyond standard approaches by including the competition of all $\sigma(\Gamma)$ and $\sigma(\Gamma^2)$ tunnel processes\cite{27, 28, 29}.

Voltage biased energy transport.—Even at zero thermal bias ($T_L = T_R = T$) the energy transport spectrum is much richer than that for the charge: Already a first glance at the charge and energy conductance plotted in Fig. 1(a) and (b), respectively, reveals a significant gain in contrast in the latter due to its many sign changes. In these plots the stage is set by resonant tunneling features which are well understood. These occur when one of the four single-electron addition energies $\varepsilon_\sigma$ and $\varepsilon_\sigma + U (\sigma = \uparrow, \downarrow)$ of the nanostructure matches $\mu_{\text{L,R}} = \pm V/2$ at one of the lines labeled (i)–(iii). Indicated by (i) are resonant tunneling transitions between the ground states of subsequent electron numbers $n$ of the dot ($0 \rightarrow 1$ and $1 \rightarrow 2$). Corresponding to the peaks in $\partial I_C/\partial V$ associated with one-electron processes $|\uparrow\rangle \rightarrow |0\rangle$ and $|\downarrow\rangle \rightarrow |2\rangle$, respectively, there are sawtooth-shaped resonances\cite{23} in $\partial I_E/\partial V$ as function of the gate voltage controlling the level position $\varepsilon$ as shown in the inset in Fig. 1(b).
sign change reflects the fact that excess energy is carried by electrons or holes. This basic energy transport feature reappears at several positions in Fig. 2(b), e.g., also when (ii) a resonant tunneling process additionally excites the dot or (iii) such a process starts off in the \( n = 1 \) excited state. The broadening of all these resonant tunneling lines is determined by the temperature for \( T \geq \Gamma \) (as in our calculation) and by \( \Gamma \) at lower temperatures.

Qualitative differences show up inside the diamond-shaped off-resonant regime—opened up by the Coulomb interaction \( U \)—where the simple resonant picture breaks down. Here coherent electron-hole processes become important as well, with the associated inelastic excitation threshold \( \Delta = \varepsilon_\uparrow - \varepsilon_\downarrow \). For voltages \( V \gtrsim \Delta \) only elastic \( \mathcal{O}(\Gamma^2) \) tunnel processes are possible which produce a smooth nonexponential background in both \( \partial I_C/\partial V \) (qualitatively similar to that found for metallic islands) \(^{(18,19)}\) and \( \partial I_E/\partial V \). Above the threshold line \( V = \Delta \), indicated by (iv) in Fig. 2(b), inelastic tunneling processes \( \mathcal{O}(\Gamma^3) \) set in, yielding the characteristic step in the charge conductance in Fig. 2(a) at \( V = \Delta \) all across the off-resonant regime. This feature is of central importance to transport spectroscopy in three-terminal \(^{(11)}\) and scanning probe \(^{(13)}\) setups. Our calculations show that the energy conductance \( \partial I_E/\partial V \) also shows such an inelastic tunneling feature at the line (iv) in Fig. 2(b). As expected, it changes sign when electron and hole processes change roles, similar to the sawtooth-shaped resonances discussed above, but now when tuning the level position through the center of the off-resonant regime (\( \varepsilon = -U/2 \)). However, a radical difference appears in the inelastic step magnitude at \( V = \Delta \) as function of the level position \( \varepsilon \) in Fig. 2(a): Whereas for the charge conductance it is smooth and featureless, the energy conductance amplitude is sharply enhanced when \( \varepsilon \approx -\Delta \) or \( \varepsilon + U \approx \Delta \). Correspondingly, the central part of the horizontal inelastic tunneling onset in Fig. 2(b) is completely missing, in contrast to Fig. 4(a). The relatively low energy current in the central region can be traced back to two effects: First, charge fluctuations are suppressed here (\( n = 1 \)) and only inelastic tunneling processes \( \langle \downarrow \rangle \leftrightarrow \langle \uparrow \rangle \) occur, both of which are \( \mathcal{O}(\Gamma) \), much smaller than the \( \mathcal{O}(\Gamma) \) main resonant features. Additionally, there is a significant partial cancellation of the positive energy current due to inelastic tunneling relaxation processes \( \langle \uparrow \rangle \to \langle \downarrow \rangle \) by the negative energy current of inelastic tunneling excitation processes \( \langle \downarrow \rangle \to \langle \uparrow \rangle \), relating to the generic electron-hole symmetry between two resonances (here at \( \varepsilon = -U/2 \)). The strong enhancement of \( \partial I_E/\partial V \) occurring when leaving this central region is remarkable: Everywhere in Fig. 2(a) we are still far from resonance in the sense of level position relative to the resonance peak width, \( |\varepsilon - \mu_L|,|\varepsilon + U - \mu_R| \gg T,\Gamma \). The increase is due to the inelastic tunneling relaxation process being overridden by a faster two-step resonant tunneling \( \mathcal{O}(\Gamma) \) relaxation, \( \langle \uparrow \rangle \to \langle \downarrow \rangle \) or \( \langle \downarrow \rangle \to \langle \uparrow \rangle \). This two-step relaxation, sketched in Fig. 2(b), involves real occupation of states \( \langle \downarrow \rangle \) and \( \langle \uparrow \rangle \) and lifts the cancellation of the signed energy currents, thereby significantly increasing it at (v) in Fig. 2(a). Such a combined process is called cotunneling assisted SET \(^{(41,43,49,50)}\) (COSET).

The above shows that combining a three-terminal setup with measurements of the energy conductance may be very promising. It provides new qualitative information about relaxation processes. If one experimentally has the full stability plot in Fig. 4(b) at hand, the above discussed regimes are separated by a sharp feature (v). This almost goes unnoticed in the charge conductance plot in Fig. 4(a) which is accessible by standard experimental techniques. Moreover, the much enhanced effect of COSET in the energy current should be experimentally accessible since the weaker COSET features in the charge transport have even been measured \(^{(26,31,111)}\). Furthermore, this indicates that the analysis of two-terminal thermoelectric measurements requires extra care. Often the two types of inelastic tunneling regimes, pure inelastic tunneling and COSET (containing the labels (iv) and (v) in

\[ \text{Fig. 2.} \quad (\text{Color online}) \ (a) \ \partial I_C/\partial V \text{ (black) and } \partial I_E/\partial V \text{ (red) at } V = \Delta \text{ as function of the level position/gate voltage. The dashed lines [also in (c)] indicate the points where } \pm V/2 \text{ equals the difference between the initial and final dot states. (b) Sketch of stability diagram indicating off-resonant processes in the various regimes. The blue arrows indicate the lines at which the cuts in (a) and (c) are taken. (c) } \partial I_C/\partial V \text{ (black) and } \partial I_E/\partial V \text{ (red) as function of the bias voltage for fixed level position } \varepsilon = -0.55U. \text{ Inset: Corresponding currents } I_C \text{ and } I_E. \]
Fig. 2(b), respectively, are not distinguished. Although for the charge conductance this may not seem to be so crucial, our results show that for the energy conductance this distinction is absolutely vital. Theoretical descriptions may fail badly for the energy current when they eliminate charge fluctuations and include only pure inelastic tunneling, even though they may seem to provide a reasonable answer for the nonlinear charge conductance similar to that in Fig. 2(a). Only in the triangular regime in Fig. 1(b) containing the label (iv) this is not important. Such approaches result in an effective model including effective exchange and potential scattering terms and are commonly used to model scanning probe experiments and quantum dots tuned deep into the Coulomb blockade regime. By adopting such a model one will fail from the start to describe the thermoelectric transport features that we identified here.

There is a second regime where the behavior of the energy conductance radically deviates from that of the charge conductance. This is seen in the bias dependence plotted in Fig. 2(c). As the inset indicates, after the large energy current enhancement at line (v) discussed above the energy current drops dramatically at lines (vi), corresponding to a strong negative differential energy conductance $\partial I_E / \partial V$ for positive energy current, exceeding the feature (v) in magnitude by far. In Fig. 1(b) this suppression occurs in the diamond-shaped region containing the label (vi). Note that the charge conductance in Fig. 2(c) is featureless there. As in the pure inelastic tunneling regime [triangle containing the label (iv)] the energy current is again suppressed, which is remarkable since now both $\mathcal{O}(\Gamma)$ relaxation pathways are turned on. However, in contrast to the regime containing the label (v), where one such pathway is active, this now leads to an energy current decrease. The reason for this is another cancellation of the leading contributions in the energy current.

We complete the classification of nonlinear energy-transport spectroscopy by noting the effect of $\mathcal{O}(\Gamma^2)$ tunneling of pairs of electrons/holes, despite the repulsive interaction. Also this feature along the line (vii) appears more prominently in $\partial I_E / \partial V$ than it does in $\partial I_C / \partial V$, positioned characteristically parallel to and midway between subsequent resonant tunneling resonances.

**Finite voltage and thermal bias.**—We finally turn to the case of a joint voltage ($\mu_L > \mu_R$) and thermal bias ($T_L < T_R$) studied in the prior work only to $\mathcal{O}(\Gamma)$. The energy conductance is shown in Fig. 3. Besides the expected asymmetry in the thermal broadening of the resonant tunneling lines associated with the different reservoirs, there are temperature-dependent offsets in both COSET lines that are highlighted in the frame. The largest offset between the blue lines splits proportional to the temperature $T_R$ and can be used experimentally to detect a temperature gradient in situ and more accurately than from the difference of resonant tunneling line broadenings alone. This cannot be done with the charge conductance (not shown).

We have also analyzed the influence of a junction and spin dependence of the tunnel constants $\Gamma_{\sigma \sigma}$. This breaking of symmetries causes COSET features to also appear in the charge conductance at the lines (vi). However, the energy conductance is always far more pronounced here and corresponds to a decrease of the energy current magnitude, in contrast to the charge current which is typically enhanced. This illustrates that transport of energy and charge through a correlated nanostructure can be quite distinct.

**Discussion.**—We have demonstrated the critical importance of the interplay of relaxation by first-order resonant tunneling and second-order inelastic tunneling for energy transport, not covered by existing thermoelectric master equations (limited to only first-order processes) nor by effective exchange scattering pictures (which eliminate first-order processes). Already accounting for a single excitation $\Delta$ turns out to restrict the applicability of the exchange scattering picture, and including more excitations further narrows it. This implies that interesting nonlinear energy transport, e.g., involving many vibrations (nano-electromechanical systems) or various spin excitations (spin caloritronics), requires careful consideration. One needs a physical model accounting for charge fluctuations and an nonequilibrium approach that includes at least the first two leading-orders of tunnel processes. Experimentally stronger couplings than those considered here may be important and interaction-induced renormalization effects may further enhance the inelastic tunneling in an interesting way.

We thank C. Cuevas, F. Haupt, M. Leijnse, and P. Reddy for valuable discussions. NMG, CBMH and DS thank the Institute for Theory of Statistical Physics, RWTH Aachen University, where substantial parts of this work have been performed. This work is part of the D-ITP consortium, a program of the Netherlands Organi-
sation for Scientific Research (NWO) which is funded by the Dutch Ministry of Education, Culture and Science (OCW). CBMH and DS were supported by the German Research Foundation (DFG) through the Emmy-Noether Program under SCHU 2333-2/1.

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