Peltier effect in strongly driven quantum wires

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(Received 3 December 2013; revised manuscript received 24 July 2014; published 14 August 2014)

We study a microscopic model of a thermocouple device with two connected correlated quantum wires driven by a constant electric field. In such a closed system we follow the time and position dependence of the entropy density using the concept of the reduced density matrix. At weak driving, the initial changes of the entropy at the junctions can be described by the linear Peltier response. At longer times the quasiequilibrium situation is reached with well defined local temperatures which increase due to an overall Joule heating. On the other hand, a strong electric field induces a nontrivial nonlinear thermoelectric response, e.g., the Bloch oscillations of the energy current. Moreover, we show for the doped Mott insulators that strong driving can reverse the Peltier effect.

DOI: 10.1103/PhysRevB.90.075124 PACS number(s): 72.10.Bg, 05.70.Ln, 71.27.+a

The physics beyond the linear response (LR) regime has recently become accessible to experimental techniques like ultrafast time-resolved spectroscopy of solids [1–8] or measurements of relaxation processes in ultracold atoms driven far from equilibrium. Most of the theoretical studies on transport beyond LR focus on charge currents driven by strong electromagnetic fields [9–23] or heat/spin transport in electric insulators subject to a large temperature gradient [24]. The thermoelectric phenomena beyond LR while important for power generation or cooling applications remain mainly unexplored, except for the specific case of noninteracting particles [25]. First efforts in filling this gap have recently been reported in [26–28] for quantum dots and mesoscopic systems, respectively.

A thermoelectric couple (TEC) is the circuit built out of two different wires and is the basic device for heat-to-current conversion or heat pumping. In this article we solve the time-dependent Schrödinger equation for a simple quantum model of TEC. It connects two wires of different materials with charge carriers being electrons and holes, respectively. Since the system is decoupled from any heat or particle reservoirs the electric field is introduced via induction. The essential tool to investigate the local thermal properties is the concept of reduced density matrix (DM) of small subsystems, enabling the study of how the entropy density increases/decreases in various parts of the TEC. It allows us also to specify the regime of the local equilibrium (LoE), when the time- and position-dependent temperature consistent with a canonical ensemble can be introduced. We show that LoE persists up to strong fields far beyond the limits of LR. Breakdown of LoE is marked by the Bloch oscillations of the particle and energy currents but also by oscillations of the particle and energy densities. We also demonstrate that in doped Mott insulators, sufficiently strong F may reverse the sign of the charge carriers.

We choose as the simple model for TEC the one-dimensional (1D) ring with L sites and spinless but interacting fermions where different materials are modeled by site-dependent local potentials ε_i. Steadily increasing magnetic flux $\phi(t)$ induces an electric field $F = -\dot{\phi}(t)/L$, as described by the time-dependent Hamiltonian [29–32]

$$H(t) = -t_0 \sum_i \{e^{i\phi(t)/L}\, c^\dagger_{i+1} c_i + \text{H.c.}\} + \sum_i \varepsilon_i n_i + V \sum_i \tilde{n}_i \tilde{n}_{i+1} + W \sum_i \tilde{n}_i \tilde{n}_{i+2}, \quad (1)$$

where $n_i = c^\dagger_i c_i$ and $\tilde{n}_i = n_i - 1/2$, t_0 is the hopping integral and periodic boundary conditions are used. V and W are repulsive interactions on nearest neighbors and next to nearest neighbors, respectively. The reason behind introducing W is to stay away from the integrable case ($W = 0$, $\varepsilon_i = \text{const}$), which shows anomalous relaxation [16,33–35] and charge transport [14,15,24,36–39]. This model allows insight into selected thermoelectric phenomena at arbitrary F directly from the solution of the Schrödinger equation, i.e., without introducing additional and possibly unjustified assumptions. At the same time, Eq. (1) captures the main phases of generic interacting systems, like metals or Mott insulators. Towards the end of this article we study the doped Mott insulators which are promising for the thermoelectric applications [40,41], while their thermoelectric response can still be described within a model of spinless charge carriers [42].

The considered TEC is not coupled to any external particle/heat reservoirs and evolves unitarily. Clearly TEC is subjected to external F and therefore not conserving energy. The Umklapp scattering processes are important for relaxation but are alone insufficient to cause thermalization [33] or to break the conservation of energy. Here the dynamics of TEC is studied within a procedure described in Refs. [14,15]. Initially $F = 0$ and we generate a microcanonical state $|\Psi(0)\rangle$ for the target energy $E_0 = \langle\Psi(0)|H(0)|\Psi(0)\rangle$ and small energy uncertainty $\delta^2 E_0 = \langle\Psi(0)|[H(0) - E_0]^2|\Psi(0)\rangle$. Then the driving is switched on and the time evolution $|\Psi(0)\rangle \rightarrow |\Psi(t)\rangle$ is calculated by the Lanczos propagation method [43] applied to small time intervals $(t, t + \delta t)$. We use units in which $\hbar = k_B = t_0 = 1$.

We model different wires of TEC assuming a symmetric situation shown in Fig. 1(a), i.e., $\varepsilon_i = -\varepsilon_0$ and ε_0 for $i \leqslant L/2$ and $i > L/2$, respectively, while the overall system is half-

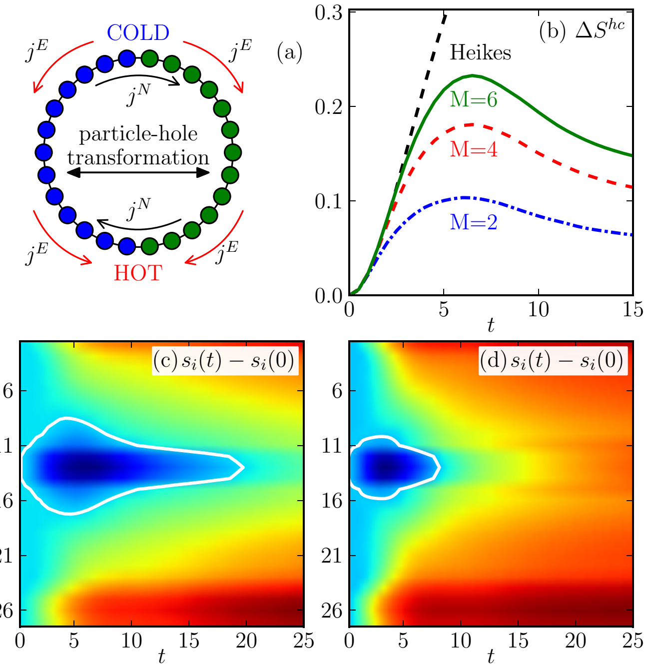

FIG. 1. (Color online) (a) Sketch of TEC. (b) Entropy difference between hot and cold junctions ΔS^{hc} for $\varepsilon_0 = 1.2$ calculated for M-site subsystems in comparison with Heikes Eq. (3). (c) and (d) $s_i(t) - s_i(0)$ for $F = 0.2$ and $F = 0.4$, respectively, for $M = 4$, $\varepsilon_0 = 1.6$, with line denoting $s_i(t) = s_i(0)$.

filled, i.e., the number of electrons $N_e = L/2$. Such a choice means that carriers in both wires are of opposite character, i.e., they are electrons and holes, respectively. The two parts of the TEC can be transformed to each other by a particle-hole transformation $c_i \to a_i = (-1)^i c^\dagger_{L+1-i}$ (see Fig. 1). As a result, the concentration of fermions on one side of each junction is the same as the concentration of holes on other side $\langle n_i \rangle = \langle a^\dagger_i a_i \rangle = 1 - \langle n_{L+1-i} \rangle$. It holds at any time provided that the same holds for $|\Psi(0)\rangle$. The Hamiltonian is invariant also under the transformation $c_i \to c_{L/2+1-i}, \phi \to -\phi$, which explains why the hot and cold junctions can be swapped by reversing $\phi(t)$ (equivalent to reversing F).

The basic characteristics of the driven TEC are obtained from charge current $j^N_i = \langle J^N_i \rangle$ and energy current $j^E_i = \langle J^E_i \rangle$. The currents are defined by the relations $\nabla J^N_i = i[n_i, H]$ and $\nabla J^E_i = i[h_i, H]$, where $H = \sum_i h_i$ and $\nabla J_i \equiv J_{i+1} - J_i$ [44]. So defined currents fulfill the continuity relations [14]

$$\frac{d}{dt}\langle n_i \rangle + \nabla j^N_i = 0, \quad \frac{d}{dt}\langle h_i \rangle + \nabla j^E_i = F(t) j^N_i. \quad (2)$$

Due to the imposed particle-hole symmetry, the charge currents are the same on both sides of the junctions $j^N_i = j^N_{L+1-i}$, while the energy currents flow in the opposite directions $j^E_i = -j^E_{L+1-i}$ [see Fig. 1(a)]. The latter property implies that the magnitude of ∇j^E_i is particularly large at the junctions, what is the essence of the Peltier heating or cooling. The energy density changes also due to the Joule heating, as represented by the source term on the right-hand side of Eq. (2). However, the heating is of the order of at least F^2 while $\nabla j^E_i \propto F$.

Since TEC undergoes a unitary evolution it stays in a pure state $|\Psi(t)\rangle\langle\Psi(t)|$ and the von Neumann entropy is identically zero. However, employing the concept of local reduced DM [45] the entropy density can be obtained from DM of small subsystems of the TEC. For subsystems of M consecutive lattice sites we calculate $\rho = \mathrm{Tr}_{L-M}|\Psi(t)\rangle\langle\Psi(t)|$ where the partial trace is taken over the remaining $L - M$ sites. Then, $S_i(t) = -\mathrm{Tr}_M(\rho \log \rho)$ is the local entropy and $s_i(t) = S_i(t)/M$ corresponding entropy density where i labels the position of the subsystem within TEC. s is a thermodynamically relevant intensive quantity [45,46] except for the low-energy regime where typically $s \propto M^{-1}$ according to the area laws [47]. Hence, we choose in this study the initial microcanonical states corresponding to high temperatures, i.e., initial $\beta(0) \simeq 0.3$. Furthermore, we also set the size to the largest available within our numerical approach $L = 26$, while the finite size effects are discussed in the Supplementary Material [48].

In order to identify the hallmarks of LoE we focus on the weak-field regime. We consider the metallic regime $V = 1.4, W = 1$ where the linear response functions are featureless [36]. Figures 1(c) and 1(d) show $s_i(t)$ for the TEC driven by $F =$ const. Major changes of $s_i(t)$ are clearly visible at the junctions, i.e., at $i = 13$ and 26. For short times $t < 10$, $s_{13}(t)$ strongly decreases (we dub it the cold junction), while $s_{26}(t)$ strongly increases (hot junction). Due to the particle-hole symmetry, $\langle n_i \rangle + \langle n_{L+1-i} \rangle = 1$ holds true to all times, hence the average concentration of fermions in subsystems covering equal number of sites from both wires constantly equals 1/2. Therefore, the change of the entropy at the junctions must be due to genuine heating/cooling. Further support for this interpretation follows from Fig. 1(b), which shows the difference of the total entropies of subsystems which cover the hot and the cold junctions. Initially, the results are independent of M, indicating that entropy is gained/lost mostly at the junctions consistently with Peltier heating $\dot{Q} = T\dot{S} = 2\Pi j^N$. At high T we can employ the Heikes formula for the Peltier coefficient in each wire $\Pi \simeq -\mu \sim \pm\varepsilon_0$. Then, the difference of entropies is

$$\Delta S^{hc} \equiv S^{\mathrm{hot}}(t) - S^{\mathrm{cold}}(t) \simeq 4\beta(0) \int_0^t dt' \varepsilon_0 j^N(t'), \quad (3)$$

while $\Delta s^{hc} = \Delta S^{hc}/M$. In the investigated regime the particle currents are determined by LR [14,15]. Hence, the rate of the entropy gain/loss at the junctions is roughly proportional to F being well consistent with the standard LR. In the Supplementary Material [48] we provide further support for the LR in the short-time regime.

An important property of the long-time regime can be inferred from Fig. 2(a) that shows j^N and j^E in the middle of the left part of TEC (far from the junctions). Initially, both currents show similar time dependence, however j^E vanishes for $t > 10$ while j^N remains large. In order to explain this nonlinear effect we recall that in the LoE regime both currents are driven by two independent forces: F and $\nabla\beta$. A particular combination of these forces may cause vanishing of j^N (Seebeck effect) or j^E (present case). In order to explicitly show that vanishing of j^E originates from compensation of two forces we instantaneously switch off one of them: the electric

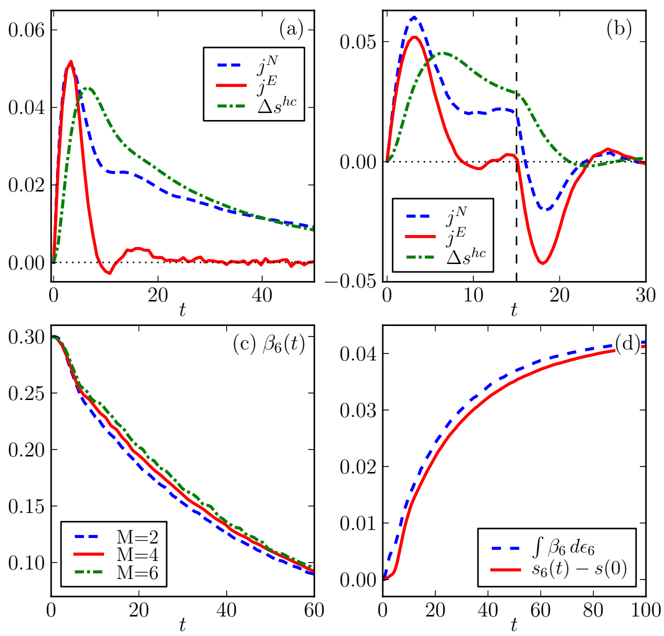

FIG. 2. (Color online) Results for $M = 4$, $F = 0.2$ and $\varepsilon_0 = 1.2$. (a) j^N, j^E in the middle of the left wire together with Δs^{hc}. (b) The same but for F switched off at $t = 15$. (c) $\beta_i(t)$ for $i = 6$, i.e., away from junctions. (d) $s_6(t) - s_6(0)$ determined directly from RDM and from $\int d\epsilon_6 \beta_6$.

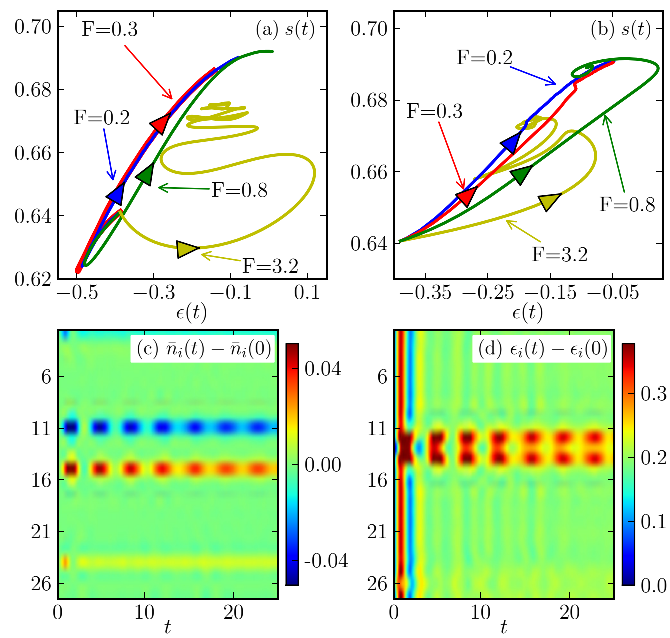

FIG. 3. (Color online) Results for $M = 4$ and $\varepsilon_0 = 1.2$. (a) and (b) Parametric plots $s_i(t)$ vs $\epsilon_i(t)$ for cold and hot junctions, respectively. (c) and (d) Oscillations of the particle and energy densities, respectively, for $F = 3.2$.

field. As shown in Fig. 2(b), the remaining force drives j^E in the opposite direction. The particle-hole symmetry implies that either j^E vanishes or $\nabla j^E \propto j^E$ is large at the junctions. As follows from Eq. (2), the latter possibility would preclude the quasistationary evolution of an *isolated* TEC. In case of open systems we expect only partial compensation of driving forces in a stationary state which diminishes the efficiency of heat pumping under a strong driving.

It has been shown for a driven homogeneous wire that ρ is block diagonal with respect to the number of particles in the subsystem. In the quasiequilibrium (QE) regime $\rho \propto \exp[-\beta(t)H_{\text{eff}}]$ within each block [45] and the spectrum $\{E_m\}$ of the effective Hamiltonian H_{eff} is independent of β. Although for small subsystems H_{eff} may significantly differ from H, one may still estimate $\beta(t)$ without specifying an explicit form of H_{eff}. For the initial microcanonical state with known inverse temperature $\beta(0)$ we determine the eigenvalues $\tilde{\lambda}_m$ of the largest block of ρ. Then a similar spectrum λ_m is determined for a driven system in a QE. Assuming the same $\{E_m\}$ one can then estimate $\beta(t)/\beta(0) = \log(\lambda_m/\lambda_1)/\log(\tilde{\lambda}_m/\tilde{\lambda}_1)$. Figure 2(c) shows the resulting $\beta_i(t)$ (averaged over $m \neq 1$) for the subsystem in the middle between hot and cold junctions. Being almost independent of M, β is a well defined intensive quantity. Finally, we demonstrate that β is consistent with the second law of thermodynamics. In Fig. 2(d) we compare $s_i(t) - s_i(0)$ determined directly from ρ with the integral $\int_0^t d\epsilon_i(t')\beta_i(t')$, where $\epsilon_i(t) = \langle h_i(t)\rangle$ is the energy density in the subsystem. Both quantities are very close to each other. Therefore, we conclude that in the QE regime one may introduce $\beta_i(t)$ consistent with the canonical ensemble as well as with equilibrium thermodynamics. This consistency breaks down only for a subsystem covering one of the junctions. In the Supplementary Material [48] we discuss $\beta_i(t)$ in more detail. We refer to Ref. [49] for a method of introducing β_i in weakly driven open systems.

Next we concentrate on nonequilibrium phenomena related with the operation of the TEC under strong F. The first one concerns the magnitude of F which destroys the LoE. Since the TEC is spatially inhomogeneous LoE can be destroyed in certain parts of TEC while persisting in the other parts. In the LoE regime, intensive quantities including $s_i(t)$ and $\epsilon_i(t)$ are uniquely determined by $\beta_i(t)$. Such a universal relation is confirmed for $F \leqslant 0.4$ in Figs. 3(a) (cold junction) and 3(b) (hot junction). In the former case the curves for weak F merge during the entire evolutions, while in the latter case it happens only in the long-time regime after the nonequilibrium transient. Results for $s_i(t)$ within the wires (not shown) are intermediate to the cases shown in Figs. 3(a) and 3(b). Hence, one can observe that the LoE regime is broken first at the hot junction. This observation could be compared to the results on the interplay between disorder and equilibration [50]. For large F, $\epsilon_i(t)$ starts to oscillate, while oscillations of $s_i(t)$ are rather limited. Therefore, the equilibrium relation between ϵ_i and s_i is broken when the *energy current* $j_i^E(t)$ starts to undergo the Bloch oscillations [32]. Note that in the homogeneous systems j_i^N and j_i^E may Bloch oscillate [10,14,19,51], still the density of particles and the density of energy do not show any oscillatory behavior. However, the TEC is inherently inhomogeneous, hence the oscillating currents imply oscillations of ∇j_i^N and ∇j_i^E. Then, according to the continuity equations (2) also $\langle n_i\rangle$ and $\langle h_i\rangle$ must undergo the Bloch oscillations which are particularly strong at the junctions and decay inside the wires as it is shown in Figs. 3(c) and 3(d).

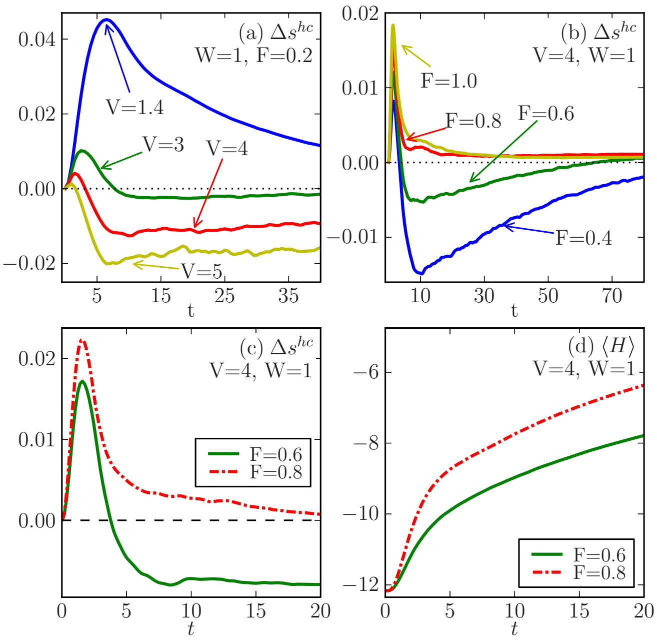

FIG. 4. (Color online) (a) and (b) $\Delta s^{hc}(t)$ for various V and F, respectively, with $M=4$ and $\varepsilon_0=1.2$. (c) and (d) $\Delta s^{hc}(t)$ and $\langle H(t)\rangle$, respectively, for $\varepsilon_0=1.6$.

Finally, we test the nonequilibrium response of TEC built out of two doped Mott insulators. Figure 4(a) shows the operation of TEC when the interaction V is tuned from small (metallic) $V<2$ to large values $V\gg 2$ corresponding, close to half-filling, to lightly doped Mott insulators. Such tuning reverses the dc flow of entropy (at longer t) and effectively interchanges the role of junctions (hot and cold, respectively). This effect is not unexpected being the result of changing the charge carriers close to half-filling from electrons in metallic regime to holes in the Mott-insulating regime. In contrast, results in Figs. 4(b) and 4(c) are surprising. One can see that under strong driving, the Mott-insulating TEC operates in the same way as expected for generic metals, i.e., the current is again carried by electrons. Breaking of the Mott insulator *ground state* by strong F has intensively been investigated during the last decade [9,11–13,52–56] and explained mostly as a kind of Landau-Zener transitions from the dispersionless ground state to a dispersionful excited state. However in the present case, the breakdown concerns a *doped* Mott insulator and involves only excited states with rather high energy. The first-guess scenario would be that it originates from a faster increase of energy at larger F. In order to exclude such a possibility, we have compared Δs^{hc} and $\langle H\rangle$ shown in Figs. 4(c) and 4(d), respectively. Despite having comparable energies (e.g., $\langle H\rangle \simeq -8$) TEC driven by $F=0.6$ and $F=0.8$ show opposite Peltier responses ruling out the effect of heating alone. Tuning the Peltier response by changing F is thus a truly nonequilibrium phenomena and its proper explanation remains a challenge.

In the Supplementary Material [48] we present general arguments explaining why not only j^N but also j^E undergoes the Bloch oscillations when F is sufficiently strong. These arguments hold for a generic tight-binding model and do not rely on the standard quasiparticle picture. Then, the oscillations of particle and energy *densities* at the TEC junctions arise directly from the continuity equations, hence they should also be generic for strongly driven TEC. We expect that rather intuitive results on doped Mott insulators are also model independent, however we are not aware of any analytical studies which could substantiate this claim. Therefore, our qualitative results for strong driving should be generic for tight-binding models.

Using the concept of reduced density matrix we have studied a model of driven TEC which offers an insight into several aspects of thermoelectric phenomena in the local equilibrium and far-from-equilibrium regimes when LR coefficients are not giving a good picture of the thermoelectric performance. In the former case (LoE) one may consistently define position-dependent temperature, which changes in time diminishing the efficiency of heat pumping. In the metallic regime of the model, strong F leads to the breakdown of the relation between local temperature $T_i(t)$ and local energy $\epsilon_i(t)$ which is incompatible with the notion of LoE. This is accompanied by fluctuations of particle and energy densities which are most pronounced in the vicinity of the junctions. Even more dramatic are the effects in the regime of doped Mott insulator where the charge carriers (within the equilibrium LR response) change the electron/hole character. Here we find that large F can even reverse the thermoelectric response, which cannot be explained by heating.

The authors acknowledge stimulating discussions with Veljko Zlatič. This work has been carried out within the project DEC-2013/09/B/ST3/01659 financed by the Polish National Science Center. P.P. acknowledges the support by the Program P1-0044 and project J1-4244 of the Slovenian Research Agency.

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