ARTICLE IN PRESS
COMREN:3340

C. R. Physique ●●● (●●●●) ●●●–●●●

Contents lists available at ScienceDirect

Comptes Rendus Physique

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Thermoelectric mesoscopic phenomena / Phénomènes thermo électriques mésoscopiques

Nonlinear phenomena in quantum thermoelectrics and heat

Phénomènes non linéaires dans le transport quantique thermoélectrique et thermique

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ARTICLE INFO

Article history:
Available online xxxx

Keywords:
Nonlinear thermoelectrics
Mesoscopic systems
Quantum heat transport

Mots-clés :
Thermoélectricité non linéaire
Systèmes mésoélectriques
Transport de la chaleur en régime quantique

ABSTRACT

We review recent developments in nonlinear quantum transport through nanostructures and mesoscopic systems driven by thermal gradients or in combination with voltage biases. Low-dimensional conductors are excellent platforms for analyzing both the thermoelectric and heat dynamics beyond the linear response because, due to their small size, a small temperature difference applied across regions gives rise to large thermal biases. We offer a theoretical discussion based on the scattering approach to highlight the differences between the linear and the nonlinear regimes of transport. We discuss recent experiments on quantum dots and molecular junctions subjected to strong temperature differences. Theoretical predictions concerning the Kondo effect and heat rectification proposals are briefly examined. An important issue is the calculation of thermoelectric efficiencies including nonlinearities. Cross Seebeck effects and nonlinear spin filtering arise in superconductors and topological insulators, while mixed noises between charge and heat currents are also considered. Finally, we provide an outlook on the possible future directions of the field.

RÉSUMÉ

Nous passons en revue les dévelopements récents relatifs au transport au travers de nanostructures et de systèmes mésoscopiques, engendré par des combinaisons de gradients de température et/ou de potentiel. Les conducteurs de basses dimensions constituent d'excellents systèmes pour étudier les dynamiques thermoélectrique et thermique au-delà de la réponse linéaire, une petite différence de température engendrant de forts gradients thermiques du fait des petites tailles. Nous présentons une théorie basée sur une approche de *scattering* pour illustrer les différences entre les régimes de transport linéaire et non linéaire. Nous discutons des expériences récentes sur des boîtes quantiques ou des jonctions moléculaires soumises à de fortes différences de température. Des prédictions théoriques relatives à l'effet Kondo et à la rectification du transport thermique sont brièvement examinées. Un point important est le calcul des rendements thermoélectriques en présence de non-linéarités. Nous coinsidérons aussi des effets Seebeck croisés avec du filtrage de spin non linéaire qui se produisent dans des supraconducteurs et des isolants

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http://dx.doi.org/10.1016/j.crhy.2016.08.005

Please cite this article in press as: D. Sánchez, R. López, Nonlinear phenomena in quantum thermoelectrics and heat, C. R. Physique (2016), http://dx.doi.org/10.1016/j.crhy.2016.08.005

topologiques, et le mélange des bruits de courant de charge et de chaleur. Finalement, nous discutons les directions futures possibles dans ce domaine.

1. Introduction

Nonlinear phenomena are common in nature. Certain effects occurring in the electronic transport through solids (diode behavior [1], negative differential resistance in tunnel junctions [2], Gunn oscillations [3]) are unique to the nonlinear regime and can be only explained within theoretical models that include a nonlinear relation between the forces and the fluxes. In the quantum realm, electronic transport is dictated by the scattering properties of the conductor. The latter is typically attached to multiple reservoirs with well-defined electrochemical potentials and temperatures deep inside the reservoirs. As a consequence of voltage or thermal shifts applied to these terminals, currents are associated with both charge and energy flow through the sample. Importantly, this is a nonequilibrium problem and therefore nonequilibrium responses must be, in general, determined. In the linear regime (first-order perturbation in the shifts), an expansion around the equilibrium point allows us to express the responses in terms of the equilibrium potential. Linear responses then obey symmetry and reciprocity relations, as demonstrated by Onsager [4]. However, beyond the linear response, one needs to find the charge buildup on the conductor induced by the external shifts, which in turn affects the sample potential. The problem thus becomes self-consistent and is generally difficult to solve. This paper presents an account of the most recent advances toward a clear understanding of the nonlinear transport properties of mesoscopic and nanoscopic systems when both driving forces (voltages and temperature biases) are active at the same time. Accordingly, it is expected that transport is dominated by thermoelectric effects and that not only charge, but also heat dynamics, play a significant role. The subject is important due to the nanostructures' ability to generate large gradients with moderate applied shifts (e.g., superlattices [5] and molecular junctions [6]) and to their impact on the quest for energy harvesters and thermoelectric coolers with improved efficiencies [7–11].

The Seebeck effect is the generation of a voltage across the conductor that counterbalances the applied temperature difference in open circuit conditions (zero net current) [12]. In the linear regime, the Seebeck coefficient or thermopower is thus given by the ratio of two responses: the thermoelectric and the electric responses. For macroscopic conductors, the thermal gradient is small and the thermopower is independent of the temperature and voltage biases. In turn, the Peltier effect is the creation of a heat flow in response to an applied voltage in isothermal conditions (zero thermal gradient). In contrast to the Joule heat, which is quadratic in voltage, the Peltier heat is reversible: it changes direction if the electric field is reversed. Due to reciprocity, both Seebeck and Peltier coefficients are nicely related to each other. Whether or not this symmetry relation holds beyond linear response is one of the issues to be tackled in a nonlinear thermoelectric transport formalism. Finally, it is worth noting that heat can be also generated with a thermal difference in the isoelectric case (no applied voltage). Up to first order in the temperature bias, the response is constant and given by the thermal conductance. This Fourier law is not generally met in the nonlinear regime, where the heat now becomes a function of higher orders in a temperature expansion, and thermal rectification effects may arise [13]. This is an important point since the thermoelectric figure of merit, which is typically written in terms of linear responses only (electric conductance, thermopower and thermal conductance), should be generalized to the nonlinear regime as a ratio between the generated power (nonlinear current times voltage) and the nonlinear heat flow. Although this obviously has many practical implications, the interest in the scientific literature has been complemented with an elucidation of the fundamentals of electronic and heat quantum transport subjected to large fields. The results obtained in the last few years demonstrate that mesoscopic conductors behave in a remarkable way when they are driven far from equilibrium with both electric and thermal means.

We begin our discussion with a simple problem. Consider a two-terminal phase-coherent conductor. (The case of an arbitrary number of leads will be treated later.) Let I (J) denote the electric charge (heat) current measured at the left (L) contact. The scattering approach yields the currents

$$I = \frac{2e}{h} \int \mathrm{d}E \, \mathcal{T}(\mathcal{U}, E)[f_\mathrm{L}(E) - f_\mathrm{R}(E)] \,, \tag{1}$$

$$J = \frac{2}{h} \int \mathrm{d}E \, (E - \mu_\mathrm{L}) \mathcal{T}(\mathcal{U}, E)[f_\mathrm{L}(E) - f_\mathrm{R}(E)] \,, \tag{2}$$

where spin degeneracy has been assumed. Here, $\mathcal{T}(\mathcal{U}, E)$ is the transmission probability for a carrier with energy E traversing the conductor with a potential landscape given by the electrostatic potential $\mathcal{U} = \mathcal{U}(V_\mathrm{L}, V_\mathrm{R}, \theta_\mathrm{L}, \theta_\mathrm{R})$ that defines the screening properties of the conductor. The potential is, quite generally, a function of position but here we consider the homogeneous case for the sake of definiteness. Crucially, $\mathcal{U}$ contains a nonequilibrium term that is a function of the applied voltages, V_L and V_R, and the temperature shifts, θ_L and θ_R. This term is determined from electron–electron interactions that govern screening effects out of equilibrium. Hence, the transmission depends on the applied driving fields and the dependence will be strongest for conductors with poor screening qualities as is usual in nanostructures. This dependence

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