Mesoscopic Thermoelectric Phenomena : Foreword

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Abstract

We place in context the articles that appear in this special issue of the Comptes Rendus de l'Académie des Sciences (Comptes Rendus Physique Vol. 17 Issue 10 December 2016) entitled *Mesoscopic Thermoelectric Phenomena*, and we briefly mention some of our contributions to the subject.

1. Introduction

Thermoelectricity is a two-way process. Either a temperature gradient across a material can produce electricity (the Seebeck effect), or an electric current through the same material can create a temperature difference between its two sides (the Peltier effect). In other words, thermoelectric effects can be used either for harvesting useful energy from wasted heat (Seebeck) or for cooling things (Peltier) without the device having any moving parts.

Wasted heat yields a huge loss of energy in many domains. This is the case in the big industries (e. g. the power stations where electricity is produced), the engines of cars, trains, ships, planes, and the big data centers. Let us consider a car: only 30% of its gas consumption is used for its motion, while 70% produces heat, typically 40% disappearing through the exhaust pipe. This has led physicists to revisit thermoelectric effects. Could we use the Seebeck effect to convert the wasted heat into useful electrical power? At smaller scales, the laptops and cell phones also generate a lot of heat, using up the energy in the batteries in such a manner that they need to be charged very often. Could mesoscopic thermoelectric effects contribute to improving the heat management in nanostructures, converting the wasted heat in an useful energy supply? If so, this could be a route to saving a significant amount of energy in numerous applications. This could provide autonomous power supplies for the medical devices (pace-makers, etc) or for the internet of things, by taking advantage of small temperature differences (such as those between the human body and its environment).

Another challenge is refrigeration, notably the cooling of hot spots in microprocessors. The last decades have been characterized by an exponential growth of the on-chip power densities. Values of the order of $100W/cm^2$ have become common [1], a power density which is similar to that emitted by the core of a nuclear reactor! For comparison, $7000W/cm^2$ characterizes the surface of the sun. Needless to say that a working chip will be damaged if one does not quickly extract the heat produced. Such problems of overheating are greatly increased by the introduction of ultra-small transistors and nanowire devices which conduct heat much less well than traditional information processing technologies (CMOS, etc). More than our ability to reduce their sizes, the limitation of the performances of today microprocessors comes mainly from the difficulty of managing heat in ever-smaller integrated circuits, notably cooling the transistor drain areas. Progress could come from a more efficient use of the Peltier effect, notably for a local cooling of the hot spots. A better understanding of the thermoelectric effects, of the phonons and of the heat and entropy at scales going from the nanometer in molecules up to a few microns in quantum dots and nanowires might give the solution for these problems of overheating.

Since a few decades, the mesoscopic physics community has studied charge transport in nanostructures, considering the effect of a voltage difference at a uniform temperature, but more recently that these investigations have been

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extended to the study of the combined effects of voltage and temperature gradients in the same nanostructures (for a very recent review see [2]). This special issue contains contributions from this community. It is not intended as a review of all current progress in thermoelectrics, instead it draws attention to certain exciting topics in the thermoelectric and thermal response in mesoscopic systems. For a reader interested in the basics of thermoelectricity in bulk semiconductor materials, we recommend the textbooks [3–5] and the reviews [6–8]. We also note that exciting progress is being made using cobaltates [9] (correlated electron systems), ionic liquids [10], new semiconductor materials and polymers. A broad overview of the recent progress in thermoelectricity, notably using oxydes, intermetallic compounds and small systems can be found in the lectures given by Antoine Georges at the College de France in 2012-2013 and 2013-2014 [11]. Other reviews on these subjects include [12–15].

Independently of the hope of making technological progresses, mesoscopic thermoelectric phenomena deserve to be studied from the point of view of fundamental physics. At a practical level, the thermoelectric response of a system gives us different information about that system than its electrical response, and this information can be crucial to understanding the physics of nanostructures. More fundamentally, the concept of heat and entropy at the nanoscale is different from in macroscopic systems, and this represents an important new domain of study. The second law of thermodynamics is at the heart of our understanding of irreversible processes in the macroscopic world. At smaller scales, this law can be violated by fluctuations. These fluctuations and the large deviations around the second law were the subject of the lectures given in 2016 by Bernard Derrida at the College de France [16]. In this special issue, the contribution of Koski and Pekkola illustrates this new trend, showing how Maxwell's demons can be realized in nanoscale electronic circuits.

In general, mesoscopic thermoelectric transport involves considering a nanostructure connecting two electron baths (the hot and cold sources) characterized by Fermi-Dirac distributions of different temperatures and chemical potentials, and measuring the induced charge and energy currents through the nanostructure. In the limit where transport becomes inelastic and comes mainly from phonon-assisted hops, the nanostructure is not only coupled to two electron baths, but also to a third bath of phonons characterized by its own Bose-Einstein distribution. In other words, the investigation should be extended to setups contacting not only two electron baths, but also a third bath being a phonon bath (see the contribution of Jiang and Imry) or a third electron bath whose thermal fluctuations are coupled capacitively to the part of the nanostructure that carries electrical currents (see the contribution of Thierschmann, Sanchez, Sothmann, Buhmann and Molenkamp). Other similar thermoelectric ratchet effects in multiterminal setups are also described in Refs. [17–19]). Multiterminal thermoelectric transport becomes nowadays an active field of research.

Experimentally, there have been works on thermoelectric phenomena in quantum systems since the beginning of the study of quantum transport through nanostructures [20, 21]. However, they were limited by the lack of accurate thermometry at the nanoscale, without which one could only perform the most basic of analysis of these phenomena. In recent years this limit has increasingly disappeared as a number of thermometry techniques are being perfected, sometimes using thermoelectric effects [22]. We are now starting to get a quantitative picture of the thermoelectric responses of various quantum systems. In the theoretical physics community, the few pioneering early works on electronic thermal transport (Enquist-Anderson [23]) or thermoelectric effects (Sivan-Imry [24]) were appreciated, but little else was done due to the lack of experiments. However, now an increasing number of theorists are actively working on such problems. In addition to interactions between theorists and experimentalists with a background in mesoscopic physics, there are increasing fruitful contacts with the community of physicists working on the thermodynamics of small systems in other contexts.

2. A brief overview of the works in this special issue

This special issue brings together the contributions from twelve groups, which review and place in context their recent works.

It begins by four theory papers. Paper 1 (Jiang and Imry, `DOI: 10.1016/j.crhy.2016.08.006`) describes inelastic thermoelectric transport in mesoscopic systems coupling a source to a drain, while also being coupled to a phonon bath. It starts from the case of linear and nonlinear transport above a barrier before discussing more generally inelastic thermoelectric transport assisted by a heat bath. Rectification and transistor effects in the nonlinear regime for inelastic transport are considered.

Paper 2 (Sánchez and López, `DOI: 10.1016/j.crhy.2016.08.005`) describes non-linear thermoelectric transport in nanostructures. This point is particularly important for many thermoelectric applications of nanostructures. The reason is that linear-response theory usually fails when the temperature drop on the scale of the electrons' relaxation length (typically given by the electron-electron or electron-phonon scattering length) is *not* small compared to the average temperature. In bulk materials the temperature drop happens over millimetres when the relaxation length is tens of nanometres, so the temperature drop on the scale of the relaxation length is tiny even when the hot and cold reservoirs have very different temperatures. In contrast, in a nanoscale thermoelectric, the hot and cold reservoirs are separated only by the nanostructure itself. In this case a large temperature difference immediately means one is outside the linear-response regime. Then the thermoelectric figure of merit, ZT, ceases to describe the thermoelectric's efficiency [25–30]. While the theoretical modelling of systems beyond the linear regime is difficult, it will certainly be important for many applications, and this paper gives crucial steps in this direction.

Paper 3 (Benenti, Ouerdane and Goupil, `DOI: 10.1016/j.crhy.2016.08.004`) shows us the interest of being close to an electronic phase transition, notably near an Anderson transition. This work opens clear perspectives for the future, in which interacting solid-state systems near phase transitions could be candidates for efficient thermoelectrics (see also [31]). This clearly presents a challenge for theoretical physicists, since such systems are among the most difficult to understand. In addition, this paper discusses the case where the electrons interact in non-integrable systems with momentum conservation, where arguments corroborated by numerical simulations show us that the Carnot efficiency is achieved at the thermodynamic limit.

Paper 4 (Lambert, Sadeghi and Al-Galiby, `DOI: 10.1016/j.crhy.2016.08.003`) gives a review of the thermoelectric properties of single molecules and porous nanoribbons. This is a crucial example of how one can calculate the transmission function of complicated molecular structures (typically using density functional theory), and thereby predict that structure's thermoelectric response. This paper reviews works in which the authors have explored how a careful choice of the molecule would allow one to engineer a transmission function which changes rapidly with energy, thereby greatly enhancing the thermoelectric response.

Paper 5 (Svilans, Leijnse and Linke, `DOI: 10.1016/j.crhy.2016.08.002`) gives a careful quantitative comparison between experiments and theories. Such a comparison is crucial for the development of better theoretical models. They describe experiments concerning low temperature thermoelectric transport in quantum dots, as one varies their properties with gates. They provide a general discussion concerning the devices and methods (energy scales, thermal bias, thermometry, thermoelectric measurement) and show how the electrical conductance g and thermopower S vary as a function of a gate voltage. They compare the results to both the predictions given by a Landauer theory and by a theory involving a single-electron tunnelling approximation.

Paper 6 (Thierschmann, Sánchez, Sothmann, Buhmann and Molenkamp, `DOI: 10.1016/j.crhy.2016.08.001`) reviews experiments on the simplest existing machine capable of converting heat into electrical power (see also similar experiments in [17, 18]). Such machines consist of a three terminal device made of a pair of Coulomb-coupled quantum dots. They are a perfect test-bed for understanding the thermodynamics of such conversion at the microscopic scale. While these first devices have low efficiencies and only work at cryostatic temperatures, there is no theoretical reason why they could not be engineered to have high efficiencies and work at room-temperature or higher. Thus they may pave the way to new types of energy harvesting. The authors also point out that this may be a route towards all thermal transistors.

Paper 7 (Narayan, Pepper and Ritchie, `DOI: 10.1016/j.crhy.2016.08.012`) illustrates how the thermoelectric response can be used as a experimental probe of a poorly understood state of matter, reviewing recent thermoelectric transport measurements through mesoscopic two-dimensional electron gases where the carrier concentration is not large enough to have a mesoscopic Fermi gas, and not low enough to have a rigid Wigner molecule. Both the electrical conductance g (which does not exhibit the usual localized behaviour while $g < 2e^2/h$) and the thermopower S (which shows large oscillations much above the value predicted by the Mott formula) exhibit spectacular and still not well understood behaviours as one varies the carrier density.

Paper 8 (Koski and Pekola, `DOI: 10.1016/j.crhy.2016.08.011`) addresses also very fundamental issues, creating Maxwell demons using single-electron tunnelling in electronic circuits. It includes the realization of the Szilard engine in a single-electron box, a single-electron refrigerator, and the realization of an autonomous demon in coupled single-electron circuits. It demonstrates also that logical operations can be performed at a cost not far from the Landauer limit $k_B T \ln 2$.

Paper 9 (Courtois, Nguyen, Winkelmann and Pekola, `DOI: 10.1016/j.crhy.2016.08.010`) is an example of another challenge for thermoelectric effects in nanostructures; the quest for micro-Kelvin refrigeration. The hope is to use thermoelectric effects to cool micron-sized structures to temperatures below 1mK. This refrigeration would enable the experimental study of low temperature phases of electronic systems (quantum phase transitions, etc) at much lower temperatures that presently. For such cooling to be feasible, one must ensure significant cooling powers, when most nanostructures to date have cooling powers in the pico-Watt regime. This paper reviews work in which the authors achieved nano-Watt cooling power using Normal metal – Insulator – Superconductor junctions, while refrigerating an electron gas down to one fifth of its surrounding's temperature (from 150 mK to 30 mK).

Paper 10 (Xiong and Volz, `DOI: 10.1016/j.crhy.2016.08.009`) and Paper 11 (Bourgeois, Tainoff, Tavakoli, Liu, Blanc, Boukhari, Barski and Hadji, `DOI: 10.1016/j.crhy.2016.08.008`) address the fact that phonons must be controlled, if one wishes to achieve efficient thermoelectric power generation or refrigeration. Phonons typically always carry heat from hot to cold in a manner that short-circuits the thermoelectric device. Thus, reducing the phononic heat transport is essential. Unfortunately it is much harder to control the flow of phonons than of electrons, to a large extend because they are chargeless. Paper 10 discusses theoretically how one can reduce the phonon mean free path by nanostructuration, opening the road towards a spectral engineering of the phonons. Paper 11 provides experimental measurements of the thermal conductance of nanowires and thin films from low temperature up to room temperature, showing us how the phonon mean free path can be reduced by constrictions, periodic structures, and nano-inclusions.

This special issue ends with Paper 12 (Grenier, Kollath and Georges, `DOI: 10.1016/j.crhy.2016.08.013`) which considers mesoscopic thermoelectric effects with cold atoms rather than electrons. They show theoretically how the Peltier effect can be used as a new cooling procedure with improved cooling power and efficiency compared to the usual evaporative cooling. Decreasing further the temperature of the atomic gases might lead to a new generation of experiments which could allow us to understand better correlated fermion systems, an issue of the highest fundamental importance nowadays.

Taken together these twelve papers show the breadth of situations in which quantum thermoelectric phenomena could be present. They also make it clear that despite the significant advances in recent years, much of the most interesting territory remains to be explored.

3. A brief overview of some of our recent works

Finally, we briefly mention some of our recent contributions to thermoelectric transport in nanostructures and give references.

Thermoelectric transport in disordered nanowires in the field effect transistor device configuration has been studied theoretically in Refs. [32–35]: the nanowires are assumed to be deposited on a substrate below which there is a gate (one dimensional MOSFETs). Applying different voltages upon the gate in the presence of a potentiel or temperature difference between the source and the drain, one can induce electron transport either in the bulk, at the edges or even outside the conduction band. The low temperature elastic regime has been considered in Ref. [32], while the inelastic activated regime taking place at higher temperatures have been studied in Ref. [33]. In the activated regime, the nanowire's electrons are localized and can mainly propagate from one localized state to an another by absorbing or emitting the phonons of the substrate (Mott variable range hopping). Near the band edges, the thermopower S becomes very large while the electrical conductance g vanishes. Measurements at room temperature of S and g using Si and Si/Ge nanowires in the field effect transistor device configuration are given in Ref. [36], showing a large enhancement of S around the band edge. To increase the efficiency, one can take large arrays of nanowires in parallel [34], since the electrical conductances add while the thermopower self-averages. The performances of such arrays have been evaluated in Ref. [34]. In Ref. [35], it has been underlined that these arrays provide also tools for a gate control of heat transfer at micron scales: If the Fermi energy of the conduction electrons is in the bottom of the conduction band, the nanowire's electrons absorb the substrate phonons near the source and remit them near the drain when a voltage bias is applied. Ratchet effects in the hopping regime of these setups have been studied in Ref. [19].

Refs. [37, 38] shows that the wave-like nature of electrons places bounds on thermoelectric's power output and efficiency, which are absent in classical thermodynamics. In any heat-engine or refrigerator, there is an upper bound (a "quantum bound") on the power output per unit cross-section. Refs. [37, 38] and [39] show how to maximize the efficiency of a thermoelectric at *given* power within a Landauer scattering approach for two-terminal and three-terminal systems respectively: in both cases one must ensure that the transmission functions act as band-pass filters in energy. It is argued that this is better for engineering applications than simply maximizing the efficiency by taking a delta-like transmission, as that gives vanishing power output. The resulting upper bound on efficiency is more stringent that of Carnot, and only comes close to the Carnot efficiency when the power output is much less than the quantum bound on power output. It is not yet clear if these bounds apply beyond scattering theory, such as when there are significant single-electron charging effects.

Ref. [40] considers non-local power generation in a four terminal quantum device. Heat flows from hot to cold in part A of the device, causing electrical power generation in part B of the device. Remarkable, the nature of the quantum system considered (Coulomb blockaded quantum dots) means that electrical power can be generated in part B of the device even when there is no energy flow on average from A to B. This "exotic" effect is shown not to violate energy conservation or the laws of thermodynamics.

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