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Thermoelectric mesoscopic phenomena/Phénomènes thermoélectriques mésoscopiques

Linear and nonlinear mesoscopic thermoelectric transport with coupling with heat baths

Transport thermoélectrique mésoscopique linéaire et non linéaire avec couplage à des sources de chaleur

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ABSTRACT

Decades of research on thermoelectrics stimulated by the fact that nano- and meso-scale thermoelectric transport could yield higher energy conversion efficiency and output power has recently uncovered a new direction on inelastic thermoelectric effects. We introduce the history, motivation, and perspectives on mesoscopic inelastic thermoelectric effects.

RÉSUMÉ

Des décades de recherche sur la thermoélectricité, stimulée par le fait que le transport thermoélectrique aux échelles nanométriques et mésoscopiques pourrait améliorer le rendement de la conversion énergétique et la puissance délivrée, ont permis de découvrir récemment une nouvelle utilisation des effets thermoélectriques mésoscopiques. Nous présentons l'histoire et les motivations des recherches sur les effets thermoélectriques mésoscopiques inélastiques, ainsi que les perspectives ouvertes par ceux-ci.

1. Introduction

Research on the science and technology of the thermoelectric phenomenon has a long history since its discovery almost two centuries ago [1]. The modern theory of thermoelectric transport in bulk semiconductors was established, with the help of energy band theory and semi-classical transport theory, in the middle of the last century [1]. The key concepts such as the figure of merit and the power factor were introduced then, which facilitated and stimulated many theoretical and experimental studies. It was found that the figure of merit (a measure of the optimal thermoelectric efficiency in a material),

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$$ZT = \frac{\sigma S^2 T}{\kappa} \tag{1}$$

is limited by the following interrelated transport quantities: the electrical conductivity σ, the Seebeck coefficient S, and the thermal conductivity κ. The latter has contributions from the electronic transport κ_e and other mechanisms (mainly from phonons in semiconducting materials, κ_p), i.e. $\kappa = \kappa_e + \kappa_p$. Increasing S usually leads to reduce the electrical conductivity σ. In addition, σ and κ_e are interrelated. In metals, these two quantities mostly follow the Wiedemann–Franz law

$$\kappa = LT\sigma \tag{2}$$

where $L \equiv a_L (k_B/e)^2$ is the Lorenz number ($a_L \sim 1$, depending on the material and the temperature). In semiconductors, similar relations are usually approximately confirmed. When the Wiedeman–Franz law holds, the thermoelectric figure of merit is approximately $S^2 e^2 / k_B^2$, which is often smaller than 1.

Mahan and Sofo proposed to improve the figure of merit by using conductors with very narrow energy bands, so that the thermal conductivity κ_e is reduced from the Wiedemann–Franz law [2]. However, this was found lately to be less effective as, for example, the output power is suppressed in the zero band width limit [3] due to, e.g., suppressed electrical conductivity [4].

Alongside with those developments, the scientific community was pursuing a better understanding of (charge) transport in nano- and meso-scale systems, localization phenomena, and strongly interacting electron systems. These studies activated research on thermoelectric transport in non-standard (including bulk) semiconductors, which is still ongoing [5]. The most influential studies are based on two seminal works by Hicks and Dresselhaus [6], who found that in reduced-dimension semiconductors, such as quantum wells (2D), quantum wires (1D) and quantum dots (0D) heterostructures, the interrelation between the electrical conductivity, the Seebeck coefficient, and the thermal conductivity can be partially decoupled. This is mainly because the density of states can be effectively enhanced and modulated (at different energies) when the spatial confinement along any direction is small enough to induce significant quantum-confinement effects. Opportunities for enhancing the thermoelectric figure of merit ZT and the power factor $S^2\sigma$ by engineering nanostructures and nanomaterials thus emerge [7–11]. In addition, when low-dimensional structures are packed up (densely) into macroscopic structures, the abundant interfaces effectively reduce the phonon heat conductivity [7,8]. If the phonon heat conductivity is reduced more significantly (e.g., by introducing effective phonon scattering centers, or packing materials with mismatched phonon impedance), then the electrical conductivity, the thermoelectric figure of merit can be improved [9]. This was demonstrated in BiTe quantum well superlattices and PbTe quantum dot superlattices [8]. In the latter, a high density of quantum dots forming a regular array induces a large electronic density of states in the PbSeTe alloy layer. Other methods, such as energy filtering and semimetal–semiconductor transition, in engineering the electronic density of states and the energy dependent conductivity, are also introduced [10,11]. The underlying physics is manifested in the Mott–Cutler formula [12,13]

$$S = \frac{\langle E-\mu \rangle}{eT}, \quad \kappa = \sigma \frac{\mathrm{Var}(E-\mu)}{e^2 T} \tag{3}$$

where $\mathrm{Var}(E-\mu) = \langle (E-\mu)^2 \rangle - \langle (E-\mu) \rangle^2$ is the variance of the transport electronic energy; the averages of powers of the electronic energy are defined as

$$\langle (E-\mu)^n \rangle = \frac{1}{\sigma} \int \mathrm{d}E \sigma(E)(E-\mu)^n [-\partial_E f_F(E)], \quad n = 1, 2 \tag{4a}$$

$$\sigma = \int \mathrm{d}E \sigma(E)[-\partial_E f_F(E)] \tag{4b}$$

Here $\sigma(E)$ is the energy-dependent conductivity and $f_F(E) = 1/[\exp(\frac{E-\mu}{k_B T}) + 1]$ is the Fermi distribution function. The energy dependence of the conductivity $\sigma(E)$ is the key quantity for engineering the thermoelectric transport properties. It can be written as $\sigma(E) = e k_B T \rho(E) b(E)$, where $\rho(E)$ is the density of states (DOS) and $b(E)$ is the energy-dependent mobility.

Nanostructures and nanocomposites have been demonstrated as effective ways to tune the carrier density of states and the energy dependence of carrier scattering, as well as to reduce the phonon thermal conductivity. These methods lead to high thermoelectric efficiency and power density for applications. Nevertheless, several nontrivial aspects of mesoscopic electron transport have not been exploited, which might give a chance to a further enhancement of thermoelectric efficiency and power. These are: (1) nonlinear effects, (2) inelastic effects (due to strong carrier–carrier interaction and carrier–phonon interaction), (3) thermoelectric transport with spatially separated electrical and thermal currents. The main purpose of this review is to emphasize those aspects and their potential values for improving thermoelectric performance, as well as the realization of thermoelectric diodes and transistors. We mainly focus on the inelastic effects and show that they can offer an alternative (new) route toward high-performance thermoelectric structures that may reduce the need for novel functional materials. We further discuss nonlinear effects and the spatial separation of heat and electrical currents in thermoelectric transport through inelastic thermoelectric transport. We will illustrate how these aspects may lead to better thermoelectrics. Several example systems are used to demonstrate the principles.

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