# S5. Constitutive Equations for Heat Conduction in Nanosystems and Non-Equilibrium Processes

**Organizers:**

- David Jou (Universitat Autònoma de Barcelona, Spain)
- Vito Antonio Cimmelli (University of Basilicata, Italy)

**Speakers:**

1. V. A. Cimmelli (University of Basilicata, Italy)
   *Entropy principle and non-local constitutive equations in nanosystems*
2. C. de Tomas (Universitat Autònoma de Barcelona, Spain)
   *The role of collective phonons in thermal transport*
3. Y. Dong (Tsinghua University, China)
   *Heat diodes and transistors: a short introduction to phononics*
4. Miroslav Grmela (École Polytechnique de Montréal, Canada)
   *Hamiltonian and nonlocal continuum mechanics*
5. Yu-Chao Hua (Tsinghua University, China)
   *The principle of least action in heat transfer process*
6. D. Jou (Universitat Autònoma de Barcelona, Spain)
   *Heat transport in nanoporous systems and heat rectification effects*
7. Orazio Muscato (University of Catania, Italy)
   *Electron Transport in Silicon NanoWires*
8. L. Restuccia (University of Messina, Italy)
   *Temperature in non-equilibrium systems with internal variables and implications on heat equation*
9. P. Rogolino (University of Messina, Italy)
   *Thermoelectric coupling in Thermomass theory*
10. L. Saluto (University of Palermo, Italy)
    *Inhomogeneous vortex tangles in superfluid helium turbulence*

11. M. Sciacca (University of Palermo, Italy)
*Effective thermal conductivity in narrow channels filled with Helium II: non-local effects and non-linear effects*

12. A. Sellitto (University of Basilicata, Italy)
*Non-local constitutive equations for thermoelectric effects and its implications on thermoelectric energy conversion*

13. F. Vázquez (Universidad Autónoma del Estado de Morelos, Mexico)
*Optimal performance and entropy generation transition from micro to nanoscaled thermoelectric layers*

# Entropy principle and non-local constitutive equations in nanosystems

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The present talk is faced with weakly nonlocal constitutive theories of heat conduction, in which the gradients of the unknown thermodynamic fields are allowed to enter the state space. Weakly nonlocal constitutive equations are especially important in nanoscale heat transfer, where heat transport equations may contain higher order gradients. As a simple illustration, we mention the Guyer-Krumhansl equation

$$\tau \partial_t \mathbf{q} + \mathbf{q} = -\lambda \nabla T + \ell^2 \left( \Delta \mathbf{q} + 2 \nabla \nabla \cdot \mathbf{q} \right), \tag{1}$$

where $\mathbf{q}$ is the heat flux, $\lambda$ is the thermal conductivity, $\tau$ the relaxation time of the heat flux and $\ell$ the mean free path of the heat carriers. The corresponding entropy and entropy flux are

$$s = s_{eq} - \frac{\tau}{2\lambda T^2} \mathbf{q} \cdot \mathbf{q}, \tag{2}$$

$$\mathbf{J} = \frac{\mathbf{q}}{T} + \frac{\ell^2}{\lambda T^2} \nabla \mathbf{q}^{\mathrm{T}} \cdot \mathbf{q}, \tag{3}$$

with $s_{eq}$ the local-equilibrium entropy, i.e. the entropy in the absence of relaxation effects. The aim of this presentation is:

- to show how the entropy principle can be applied to derive enhanced systems of transport equations describing nonlocal effects at nanoscale;
- to prove that in some physical situations nonlocal constitutive equations are necessary to explain unexpected phenomena which, at a first sight, could appear in contradiction with second law of thermodynamics.

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# The role of collective phonons in thermal transport

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A Kinetic-Collective model of phonon heat transport is presented to calculate the thermal conductivity of semiconductor materials at different size scales, from bulk systems to nanosystems. This model accounts for the role of normal (momentum-conserving) collisions in thermal transport, and provides a more accurate prediction of the thermal conductivity ([1], [2]). Within this model, the thermal conductivity is explained as a combination of a kinetic and a collective phonon heat flux with significantly different contributions. The main difference between these regimes is that, in the kinetic one, each phonon mode has its own relaxation time, while in the collective term the relaxation time is the same for all phonons. Furthermore, phonon hydrodynamics plays an important role in the collective transport, especially in nanowires ([3]). From the thermodynamic perspective this suggests the convenience of dealing separately with these two different terms, instead of considering them as a single quantity.

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# Thermal rectification based on phonon hydrodynamics and thermomass theory

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The thermal diode is the fundamental device for phononics. There are various mechanisms for thermal rectification, e.g. different temperature dependent thermal conductivity of two ends, asymmetric interfacial resistance, and nonlocal behavior of phonon transport in asymmetric structures. The phonon hydrodynamics and thermomass theory treat the heat conduction in a fluidic viewpoint. The phonon gas flowing through the media is characterized by the balance equation of momentum, like the Navier-Stokes equation for fluid mechanics. Generalized heat conduction law thereby contains the spatial acceleration (convection) term and the viscous (Laplacian) term. The viscous term combined with the slip boundary condition induces rectification if the slip factor changes with the heat flux direction. The convection term also predicts rectification because of the inertia effect, like a gas passing through anozzle or diffuser

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[3] Sellitto, A., Alvarez, F.X., Jou, D., Geometrical dependence of thermal conductivity in elliptical and rectangular nanowires, *Int. J. Heat Mass Trans.* **55** (2012), 3114–3120.

[4] Dong, Y., Cao, B. Y., Guo, Z. Y., Generalized heat conduction laws based on thermomass theory and phonon hydrodynamics, *J. Appl. Phys.* **110** (2011), 063504.

# Hamiltonian and nonlocal continuum mechanics

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Governing equations of continuum mechanics are required to be compatible with mechanics and thermodynamics. While there are important variations in the existing mathematical formulations of the compatibility with thermodynamics (called an entropy principle), the variations are significantly smaller than in the mathematical formulation of the compatibility with mechanics. A strong formulation of this compatibility, introduced by Alfred Clebsch [1] and Vladimir Arnold [2], is the following: The part of the time evolution equations that leaves the entropy unchanged is required to be Hamiltonian. In my talk I will present three results related to the Hamiltonian formulation of the classical and the extended continuum mechanics.

(1) If the Hamiltonian involves weakly nonlocal (Cahn-Hilliard type) terms then the Hamiltonian formulation implies immediately the extra (nonlocal) terms in expressions for the mass, the momentum, the energy, and the entropy fluxes [3].

(2) What is the closure that transforms the infinite (Hamiltonian) Grad hierarchy into a finite hierarchy that is also Hamiltonian? A partial answer to this question is provided. Possible appearance of dissipative terms in the mass flux in the classical fluid mechanics emerging as a reduction of the Grad hierarchy is discussed.

(3) I show that an investigation of the compatibility between an extended and the classical fluid mechanics leads necessarily to the appearance of nonlocal (Cahn-Hilliard type) terms in the thermodynamic potential.

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# The principle of least action in heat transfer process

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The principle of least action is a universal law in nature. It claims that the path (or distribution) actually followed by a physical system is that for which the action is minimized. The differential equation governing the physical phenomena can be derived via the variation of action. As for heat transfer process, Fourier's law of heat conduction describes the actual constitutive relation between temperature distribution and heat flux in most cases. Inspired by the principle of least action in mechanics, people have tried to obtain the Fourier's law by variation of a particular action. In nonequilibrium thermodynamics, the minimum entropy production rate has been regarded as the least action to derive the Fourier's law with the thermal conductivity proportional to squared temperature, which does not agree with the reality. To resolve this problem, a new quantity $UT/2$ in the dielectric material called entransy ($U$ is internal energy and T is temperature) was proposed. It is the simplified expression of thermomasss potential energy, which represents the ability of heat transfer during a time period. And, Fourier's law with constant thermal conductivity can be derived from the principle of least entransy dissipation rate. Hence, we may take the entransy dissipation rate, $-k\left(\nabla T\right)^2/2$, as the action in heat transfer process which is a measure of the irreversibility of heat transfer process not related to heat -work conversion.

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# Phonon hydrodynamic description of heat rectification in some porous silicon devices

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Heat rectification is one of the interesting features of the new developing field of research in heat transport named phononics. In analogy with electronic diodes, phononic diodes are devices aimed to rectify heat flux, i.e. the total thermal conductance in one direction is different from the conductance in the opposite direction, under the same given high and low values of temperature. Thermal diodes, as well as thermal transistors, have been possible because of the increasing control of phonon transport achieved by nanotechnology. Usually, this topic is examined on microscopic grounds, but here we use a mesoscopic approach based on phonon hydrodynamic expressions for the effective thermal conductivity of porous Si. In particular, we consider two situations: a) a device formed by a region of bulk Si and another one of porous Si, b) a device formed by a graded porous Si, i.e. a Si device whose porosity changes along the axis of the system. The expression for the thermal conductivity of porous Si we use takes into account the ratio of the phonon mean-free path to the radius of the pores. For small values of this ratio one finds classical results, with practically no rectification, whereas for high values of this ratio (small pores) the rectification ratio may achieve values of 1.3 or 1.4. We study such rectification for different porosities, different sizes of the pores, and different porosity profiles. We also consider a bulk/porous/bulk device but we show that, under the conditions analyzed in the paper, it does not act as a thermal transistor.

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[3] Criado-Sancho, M., Alvarez, F. X., Jou, D., Thermal rectification in inhomogeneous nanoporous Si devices, *J. Appl. Phys.* **114** (2013), 053512.

# Electron Transport in Silicon NanoWires

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A hydrodynamic model for silicon quantum wires is formulated by taking the moments of the multisubband Boltzmann equation, coupled to the Schrödinger-Poisson system. Explicit closure relations for the fluxes and production terms are obtained by means of the Maximum Entropy Principle of Extended Thermodynamics, including scattering of electrons with acoustic and non-polar optical phonons. By using this model, a n+ - n - n+ silicon diode has been simulated, and thermoelectric effects have been investigated.

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# Non-equilibrium temperatures in systems with internal variables and implications on heat equation

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Temperature is a concept common to all thermodynamic approaches. Different approaches may consider temperature from different perspectives. In equilibrium thermodynamics different definitions of equilibrium temperature lead to the same value. Out of equilibrium, equipartition is not to be expected and then in non-equilibrium steady states, in presence of an external energy flux, these non-equilibrium temperatures are different from each other. In this contribution, in the framework of extended irreversible thermodynamics, we investigate thermometric, caloric and entropic non-equilibrium temperatures, and the relations among them, in crystals with defects of dislocation (described by the dislocation core tensor, introduced as internal variable), when they are crossed by an external energy flux. We have in mind, for instance, the walls of a fusion nuclear reactor, which are submitted to an intense neutron flux supplied by the nuclear reaction. Furthermore, in the linear approximation, we derive the heat equation for these defective crystals. Also, in the same approximation we obtain the heat equation for piezoelectric crystals with dislocations.

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# Thermoelectric coupling in Thermomass theory

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In this talk it is proposed a nonlinear model for the thermoelectric coupling at nanoscale, in the framework of the thermomass theory [1, 2, 3] of heat conduction. It is shown that, as consequence of the non linearity of the model equations, the classical Onsager relations (OR) and the second Kelvin relation (SKR) are no longer satisfied simultaneously. The following situations are analyzed in detail:

- the OR are valid but the SKR breaks down;
- the SKR is valid but the OR breakdown;
- both the OR and the SKR breakdown.

In all these situations, the efficiency of thermoelectric coupling is evaluated. It is proved that such efficiency is strongly influenced by the nonlinear terms entering the expression of the heat flux. In order to obtain the optimal value for the thermoelectric efficiency, it is applied a new procedure based on the mathematical analysis of the local rate of entropy production along the thermoelectric process; it is assumed that the best efficiency corresponds to a minimum of the rate of energy dissipated along the process.

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# Inhomogeneous vortex tangles in superfluid helium turbulence

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Evolution of inhomogeneous vortex tangles is relevant because it may exhibit new physical effects and could clarify the role of vortices in energy transfer.

In this lecture we present two recent contributions to the description of inhomogeneous vortex tangles: in [1] we generalize previous models [3, 4] in order to describe the evolution of superfluid turbulence in counterflow situations, taking into account both nonlocal terms and inhomogeneous contributions; in [2] we consider the role of vortex diffusion on the evolution of the vortex length density.

In [1] we explore the general thermodynamic formalism leading to evolution equations that describe thermal dissipation, vortex diffusion and a new contribution to vortex formation. We discuss the meaning of Lagrange multipliers, pointing out the presence of two different contributions to the internal energy and entropy of the system. We also discuss the form of the entropy and the entropy flux, compared with those obtained in maximum-entropy formalism. Furthermore we propose an experiment in which the contribution of $\nabla L$ to the heat flux could be checked and observed.

In [2] we solve the hydrodynamic equations for radial turbulent counterflows in HeII between two concentric cylinders at different temperatures and the possibility of hysteresis in the vortex line density under cyclical variations of the heat flow (fast increase followed by a slow decrease of it) is explored.

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# Effective thermal conductivity in narrow channels filled with Helium II: laminar, turbulent, diffusive, and ballistic regimes

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Heat transport in superfluid helium has several special features related to the relative presence of phonons and rotons, the laminar or turbulent flow, and the relation between phonon mean-free path (mfp) and the radius of the container. This topic is of interest for the cooling of nanosystems. These features can be summarized in:

**Landau regime:** When the phonon mfp is short as compared with the radius $R$ of the pipe, and the heat-flux value is low enough, there is viscous laminar flow of the normal component of helium (carrying the heat flow).

**Gorter-Mellinck regime:** The laminar flow, corresponding to Landau regime, breaks down for sufficiently high values of the heat flux. In this case, quantized vortices appear and contribute to the thermal resistance,because of the frictional force between the normal component and the quantized vortices.

**Ballistic regime:** The phonon mean-free-path $\ell$ increases when temperature is lowered in such a way that for sufficiently low temperatures, it becomes comparable to (or higher than) the radius of the pipe (this happens below some 0.7 K for $R$ of the order of 0.5 mm, but it would occur for higher temperature if smaller diameters are considered)In this case, the predominant collisions are not the phonon-phonon collisions, but the phonon-walls collisions and hence the walls play a crucial role.

From a non-equilibrium thermodynamics perspective, our aim is to propose a general wide enough model able to describe the transition between these three different regimes, which are characterized by the ratios $\ell/d$, $L^{-1/2}/d$ and $L^{-1/2}\ell$ ($L$ being the vortex length density, $d$ the diameter of the tube, or the distance between the plates in the rectangular channel)[1, 2].

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# Nonlocal constitutive equations for thermoelectric effects and its implications on thermoelectric energy conversion

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Thermoelectricity is actually viewed as a very interesting source of electric power because of its ability to convert heat flow directly into electricity. In particular, thermoelectric devices as energy converters are easily scalable and do not have moving parts, or liquid fuels. These excellent features make them applicable in almost any situation where large quantities of heat tend to go to waste, from clothing to large industrial facilities. The advent of nanotechnology is widening the range of applicability of thermoelectric materials.

In the present talk, a model for coupled heat conduction by phonons and electrons is developed in the framework of Extended Irreversible Thermodynamics. Particular emphasis is given to nonlocal and nonlinear effects which may play a very important role in the conversion of heat current into electric current through thermoelectric effects.

The consequences of the nonlocal contributions on the figure-of-merit in nanowires are examined in two different situations regarding the relative values of the particles' mean-free path and the radius of nanowires. In both cases a dependence of the figure of merit on the transversal radius of the nanowire, as well as on some material coefficients related to the roughness of the walls, is pointed out. It is also shown how the figure-of-merit could be improved by controlling these coefficients.

The influence of nonlinear terms on the breakdown of the Onsager reciprocity relation between the effective transport coefficients, depending on the electric field and the temperature gradient, is analyzed as well. The maximum value of the thermoelectric efficiency is derived as a function of the figure-of-merit and of the degree of the Onsager symmetry breaking.

# Thermal performance and entropy production in nanoscaled thermoelectric layers

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In this work we address the problem of heat and electric charge transport in thermoelectric materials from the point of view of non-equilibrium thermodynamics. Due to the progress in the sub-microscaling techniques, the width of the thermoelectric element may be reduced to become comparable to the phonon mean free path (PMFP) in the system. At this scale, the heat transport regime becomes of the ballistic type and a thermodynamic description of the transition is possible if one includes memory and inertial effects. Based on generalized constitutive equations describing the time evolution of dissipative flows with thermal and electrical conductivities depending on the width of the thermoelectric element, a hyperbolic equation for temperature is obtained. This gives rise to a wave heat transport regime in the nanometric scale of lengths. The relation between dissipation and performance as well as the diffusive transport to wave propagation (ballistic) transition in the system are examined. Dissipation and performance relationship is investigated in terms of the entropy production and the so-called thermal figure of merit (TFM). We find that the wave-like behavior of temperature brings out an improvement of the cooling effect. The transition between the two transport regimes (diffusive-ballistic) is featured by a marked increasing of the TFM and the total entropy generation. An improvement of TFM in pulsed thermoelectricity is also found when going from the micro to the nanoscale of lengths.

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