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Heat transfer and thermodynamics: A foundational problem in classical thermodynamics and in contemporary non-equilibrium thermodynamics

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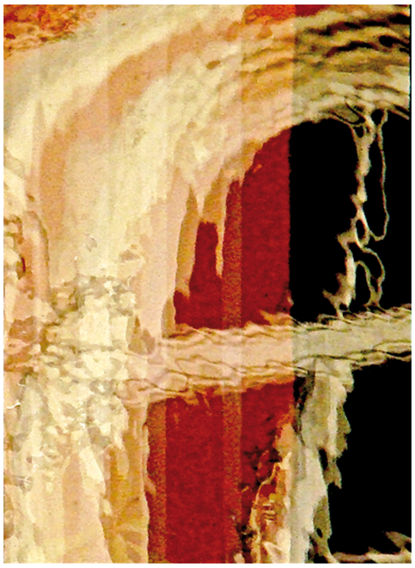

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Summary. The search for generalized heat transport equations describing Fourier's diffusive regime is a frontier in nanoscale technology and energy management, together with thermal waves, Ziman regime, phonon hydrodynamics, and ballistic heat transport, as well as their respective transitions. Here we discuss the close connection between this search and another, much less known aspect, namely, the exploration of new forms of entropy and of the second law of thermodynamics for fast and steep perturbations and high values of heat flux, which would make generalized transport equations compatible with the second law. We also draw several analogies between this situation and the confluence of Fourier and Carnot theories that resulted in a general formulation during the foundational period of thermodynamics. [**Contrib Sci** 11(2): 131-136 (2015)]

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Introduction

Research on heat transport in nanoscopic systems and the formulation of a thermodynamic framework that accommodates them are current frontiers in non-equilibrium physics. There are several analogies with the foundational period of thermodynamics in the 1820s. During that time, Jean-Baptiste Joseph Fourier (1768–1830) published the *Theorie analytique de la chaleur* (1821) [13], setting the foundations for the mathematical description of heat conduction, and Nicolas Léonard Sadi Carnot (1796–1832) published his *Réflexions sur la puissance motrice du feu et sur des machines propres à développer cette puissance* (1824) [4], which is considered the foundational cornerstone of thermodynamics. Although, at least initially, heat transport and heat engines were considered to be largely unrelated, they were soon united under the more encompassing statements of classical thermodynamics, developed in the 1850s.

Keywords: non-equilibrium thermodynamics · nanoscale heat transfer · local equilibrium · extended thermodynamics · non-equilibrium entropy

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In this article, we compare those early challenges and achievements with the current problems faced by physicists and engineers working in the field of heat transfer and thermodynamics. These problems extend beyond Fourier's law and local-equilibrium thermodynamics and we emphasize some of their conceptual aspects, especially how the search for generalized heat transport equations has been a powerful stimulus for the exploration of the foundations of thermodynamics. This effort has led to the recognition of the confluence of heat transport and thermodynamics, which for the most part had been ignored due to the dominance of more practical and urgent topics related to heat transport over subtler and more abstract aspects, such as a revised definition of entropy.

Heat transfer and thermodynamics in the early days of thermodynamics

Calorimetry and the analysis of heat transfer greatly preceded the formulation of thermodynamics. Experimental and conceptual research on specific and latent forms of heat was conducted even before the caloric theory was formulated by Antoine-Laurent de Lavoisier (1743–1794) in the *Traité élémentaire de chimie* (1789) [20]. There, he proposed a hypothetical subtle and weightless matter, the caloric, as the substance of heat, moving from higher to lower temperatures or accumulating in some hidden form in phase transitions. The caloric theory was so direct, so intuitive, and so inspiring and fruitful that it was difficult to overturn and an alternative, more realistic theory would not be proposed until the end of the 1840s.

In 1701, well before the caloric theory, Isaac Newton (1642–1727) had proposed a law of heat transfer. It stated that the rate of heat transfer between two bodies at different empirical temperatures was proportional to the difference of the temperatures. Newton's reasoning stemmed from his interest in alchemy and, in particular, in the fusion temperature of several metals and alloys. Of course, at that time this was a subject beyond the frontiers of thermometric measurement. Instead, by comparing the time that it took for a mass of material to cool from its fusion point to the environmental temperature, Newton was able to estimate, albeit indirectly and very imprecisely, the fusion temperatures he was searching for. Newton's law provided a first theoretical framework for the description of heat exchange. Incidentally, it also had an influence on Buffon's method for estimating the age of the Earth, by studying the cooling time of a hot iron sphere and then extrapolating the result to a sphere the size of our planet.

Fourier's mathematical description of heat transfer inside, rather than between different bodies was a stimulus for mathematics, physics, and philosophy. From a mathematical point of view, solving the temperature evolution equation was the motivation underlying the Fourier transformations which, despite their early rejection by some mathematicians, became a very relevant tool in mathematics and mathematical physics. In physics, they were a source of inspiration for Fick's diffusion law and Ohm's electrical transport law. They also led to extensive experimental research on the transport properties of many materials and stimulated the development of the kinetic theory of gases proposed by Rudolf Julius Emmanuel Clausius (1822–1888), James Clerk Maxwell (1831–1879), and Ludwig Edward Boltzmann (1844–1906). In the case of Boltzmann, one of the aims of his studies was to obtain detailed insights into transport coefficients. In natural philosophy, the Fourier transformations provided a mathematical elegance and rigorous framework for a paradigmatic irreversible phenomenon. They stood in contrast to Newton's mathematical framework for reversible mechanics and, especially, gravitational phenomena, which had been a paradigm of the relation between Eternal Mathematics and celestial motion.

Heat transport theory does not necessarily need nor imply an explicit and full-fledged thermodynamic framework. In fact, both Fourier and Carnot used the caloric theory, assuming the materiality of heat, but without general statements about its behavior. For instance, heat transport theory provided a dynamic equation for heat transport, one that obeyed the apparently trivial, although actually very deep condition that heat goes spontaneously from higher to lower temperatures. However, it did not state this requirement as a general law of nature, applicable to predictions beyond heat transfer. Instead, the main object of Carnot's theory, stimulated by the industrial revolution fostered by steam machines, was to explore the maximum efficiency of heat engines and the conversion of heat into work. Carnot largely ignored the rate of heat transfer, which was the problem analyzed by Fourier. Indeed, although Carnot cited the works of many scientists, he did not mention Fourier—further evidence that the problems of heat transfer and thermodynamics were considered to be unrelated.

In contrast to the rapid success of Fourier's work, Carnot's went unnoticed until it was discovered by Benoit Paul Émil Clapeyron (1799–1864) in 1835, several years after Carnot's death. In 1848 it was used by William Thomson (later Lord

Kelvin) (1824–1907) to define absolute temperature. The discovery by Julius von Mayer (1814–1878) and James Prescott Joule (1818–1889) that heat is not a material but a form of energy exchange led to the formulation of the two general laws of thermodynamics, in 1850. In that year, Clausius and Kelvin formulated their respective versions of the second law in terms of the framework of heat engines established by Carnot, but without his assumption of heat conservation. The results of both Carnot and Fourier survived this deep conceptual change, but Carnot's theory was to be reformulated. With Clausius' statement of the second law, heat transfer became an essential topic of thermodynamics, not from the point of view of the rate of exchange, but regarding the directionality of the exchange.

Half a century after Fourier's law on heat conduction, Josef Stefan (1835–1893) proposed an equation for heat radiation that was complemented by Boltzmann's description of the relation between this law and Maxwell's electromagnetic theory. Radiative heat exchange became a topic of interest in astrophysics, metallurgy, and electric bulbs. By 1900, it had led to quantum theory. Since then, heat transport, in its different forms (conduction, convection, and radiation), has been a classical topic in physics, engineering, geophysics, meteorology, and the life sciences.

Heat transport beyond a diffusive regime: Revolutions in transport theory

In the last two decades of the 20th century, the field of heat transport underwent a genuine revolution, with enlarged domains of applicability and the appreciation of new regimes and phenomena, where Fourier's theory is no longer applicable. From a microscopic perspective, Fourier's law is valid in the diffusive regime, i.e., when there are many collisions between heat carriers, but not when the frequency of collisions between heat carriers and the boundaries of the container become comparable to or higher than the frequencies of the collisions amongst the heat carriers themselves. The domain of validity of Fourier's law is described by the so-called Knudsen number, defined as the ratio between the mean free path of the heat carriers and the characteristic size of the system. When the Knudsen number is very small, collisions amongst particles dominate, the regime is diffusive, and Fourier's law is valid. When the Knudsen number is >1, the regime is ballistic, i.e., the particles move between opposite boundaries without experiencing collisions with other particles. At the beginning of the 20th century there was great interest in transport theory for rarefied gases, a topic in which the contributions of Martin Knudsen (1871–1949) were especially relevant.

The topic of transport theory developed, in part, as an extension of the kinetic theory of gases in rarefied situations. In the 1950s and 1960s, rapid advances in astronautics revived interest in rarefied gases because a relevant part of the re-entry of satellites takes place in rarefied regions of the atmosphere. In this case, the usual hydrodynamics—valid, like Fourier's equation, when collisions amongst particles have a dominant effect, namely, for small values of the Knudsen number—are no longer applicable. An extreme situation of very rarefied gases occurs in mechanics, because the effects of the collisions of particles with an object are such that each collision may be considered a single mechanical event. In the 1940s, heat transfer theory was stimulated by the observation of second sound in superfluid liquid helium. The wave propagation nature of heat, instead of the usual diffusive pattern, came as a surprise, but it did not deeply influence heat transport theory because it was considered a peculiar behavior restricted to a special physical system characterized by macroscopic coherent quantum properties. The problem of thermal waves was put in a more general perspective, albeit merely a theoretical one, by the work of Carlo Cattaneo (1911–1979) and Pierre Vernotte (1898–1970) at the end of the 1940s. Both were searching for a finite speed of propagation for thermal pulses or high-frequency thermal waves [7,8,19,34,35,40]. In the 1990s, nanotechnology emerged as a technological frontier, with a huge economic impact. In many nanoscopic systems, especially at low temperatures, the size of the system is of the order of the phonon mean free path, or even smaller, and, in some cases it is smaller than electron mean free path. In these situations, the classical transport equations for heat, electricity, and thermoelectricity are no longer valid. Usually, the situation in nanosystems lies somewhere between diffusive and ballistic, which adds to the complexity of the problem.

In considering heat transport in nanosystems, several non-Fourier regimes must be taken into account. On the one hand, even in the case of Fourier's law, thermal conductivity is no longer a purely material property; rather, it also depends on the size of the system (the radius of nanowires or the thickness of thin layers, for instance) [6,15,18,38]. Furthermore, there are typical non-Fourier regimes: heat waves, the Ziman regime, phonon hydrodynamics, and ballistic transport. These are usually dealt with from microscopic perspectives, based on kinetic equations, or by *ab initio* computer simulations. With respect to transport theory, one aim is to formulate generalized transport equations at a mesoscopic level, such that the different, above-mentioned regimes can be described by a single equation [2,3,10,11,24,27].

Heat transport in nanosystems plays a role in three contemporary industrial revolutions. Ones of them is the miniaturization leading to more powerful computers, but also to the need for computer refrigeration because of the large amount of heat that is dissipated in tiny spaces as many miniaturized devices accumulate; however, refrigeration becomes more difficult at miniaturized scales because the effective thermal conductivity is reduced with respect to the bulk values. A second revolution is in energy management, basically through photovoltaic and thermoelectric effects, which may be more efficient at nanoscales than in bulk. The third revolution is in material sciences, in which nanostructures such as superlattices, carbon nanotubes, graphene, nanoporous materials, and silicon nanowires are expected to play relevant roles in heat transport, whether for insulation and refrigeration or for delicate phonon control in heat rectification and thermal transistors and commuters in the emerging area of phononics [18].

Generalized heat transport and the frontiers of thermodynamics: Temperature and entropy

In general, heat transport is approached mainly as a mathematical theory, with a dynamic equation whose mathematical solutions are obtained under certain boundary and initial conditions, independent of thermodynamic considerations, and then compared to observations. However, when dealing with generalized equations for heat transport, a generalization of thermodynamics cannot truly be avoided because of two problems: the physical meaning of temperature in fast processes, small systems, and far from equilibrium, and the definition of entropy and the statement of the second law of thermodynamics.

Zeroth principle and temperature

Out of equilibrium, the zeroth principle of thermodynamics is no longer valid, neither in general nor in the particular, but it is relevant in the case of steady states. Intuitively, it is easy to understand that, since far from equilibrium no energy equipartition is expected, all the theoretical or operational definitions or measurements of temperature involving the interaction with different sets of degrees of freedom will lead to different values of the corresponding temperature.

Thus, for instance, the kinetic temperature may depend on the direction, the average potential energy, or other definitions. Since different degrees of freedom may have different temperatures, their respective contribution to the heat flux may differ; it could even be that for some degrees of freedom heat flux goes, as usual, from a higher to a lower temperature whereas for other degrees of freedom heat flux is in the opposite direction [5,14,21,31]. Because different measurement methods explore different aspects of the system, we must be aware of the deep meaning of temperature obtained by each one.

Second principle and entropy

One of the most fruitful and versatile statements of the second law is in terms of entropy: the entropy of the final equilibrium state should be equal to or higher than the entropy of the initial equilibrium state, after some internal constraints on the system are removed. This statement considers only the entropy of the initial and final equilibrium states, but does not refer to what happens during the intervening process. This is not a trivial issue because classical entropy is a function of state defined only for equilibrium states [23].

Attempts to generalize thermodynamics to non-equilibrium states usually invoke the local-equilibrium hypothesis, assuming that locally—in sufficiently small regions—the system is in thermodynamic equilibrium, although globally it may be far from equilibrium, by having, for instance, strong temperature or pressure gradients. This assumption allows entropy to be defined at a local level. From the balance equation for the local-equilibrium entropy, and assuming the local-equilibrium version of the entropy flux, an expression is obtained for local entropy production per unit time and volume. The second law is thus stated in terms of the positive definite character of entropy production.

This statement is clearly more restrictive than the classical statement of the second law because it requires not only that the final entropy is greater than or equal to the initial one, but also that entropy always increases, at any time and in any volume. This is incompatible with equations allowing for thermal waves, in which heat may flow during short periods from lower to higher temperature, implying a negative entropy production, or in equations of phonon hydrodynamics, in which axial heat transport may lead to heat transport from lower to higher temperatures in small regions, i.e., those comparable in size to the phonon mean free path.

Thus, generalized heat transport equations, even if they successfully describe experimental observations, may in some aspects be at odds with the local-equilibrium formulation of the second law. In this case there are several possibilities. One is to work with the integrated local-entropy production, which requires that only its integral from the initial to the final state is positive. Another is to define the generalized entropies whose positive production is compatible with generalized heat transfer equations. The advantage of the first formulation is that it does not introduce new restrictions on the equations beyond those of classical equilibrium thermodynamics; thus, while no new hypotheses are made, less information is gained about the system. The advantage of the second one is that it strives for more detailed versions of heat transport and of entropy and for insights into the relation between macroscopic and microscopic approaches.

Several possibilities to describe systems beyond local equilibrium have been examined [23]. In particular, our group at the Autonomous University of Barcelona (Universitat Autònoma de Barcelona, UAB) has been working in so-called extended thermodynamics, in which several fluxes intervening in the system are considered as additional independent variables (heat flux, diffusion flux, electric current, viscous pressure tensor, and corresponding higher-order fluxes). Both the entropy and the entropy flux depend on all fluxes, in addition to classical variables [1,12,16–18,28,33].

The generalized transport equations are the evolution equations of the fluxes, which are subject to the conjecture that local generalized entropy production must be positive at any time and at any point. The thermodynamic formulation may be carried out as an extension of either classical irreversible thermodynamics (in extended irreversible thermodynamics) [16,18,21] or rational thermodynamics (in rational extended thermodynamics) [28,37]. The ensuing generalized transport equations for heat flux (and for other fluxes) are able to address the different regimes of heat transport, i.e., heat waves, the Ziman regime, phonon hydrodynamics, and the ballistic regime, as well as intermediate regimes, without violating the tentative requirement of positive entropy production.

The main results in this field can be summarized as follows: (i) the relaxational terms in the transport equations (namely, terms in the first-order time derivative of the fluxes) correspond to second-order contributions of the respective fluxes to the extended entropy; (ii) the presence of non-local terms (namely, terms in the Laplacians or gradients of the fluxes) is related to the second-order contributions of the fluxes to the entropy flux; (iii) the corresponding contributions to the Gibbs equation of the extended entropy have the form of an intensive quantity times the differential of an extensive one; the intensive one may be compared to the Legendre multipliers used in information-theory approaches (or similar ones) to statistical descriptions of non-equilibrium steady states; (iv) the combination of linearized equations for higher-order fluxes leads to a hierarchy of equations yielding a continued-fraction expansion of the thermal conductivity in terms of the wave vector or the Knudsen number, which describes the transition from diffusive to ballistic regimes [2,9,22,36]; (v) the analysis of higher-order fluxes allows for a multilevel mesoscopic description, and thus for studies of the effects of the elimination of a set of fast variables in order to project the dynamics on slower variables, depending on the time rate of the perturbations or experiments [18,29]; and (vi) fluctuations of the fluxes around equilibrium or non-equilibrium steady state are described by the second differential of the generalized entropy.

Conclusions

The search for generalized heat transport equations (and other transport equations) is not only driven by a practical need to improve material engineering and energy management, but has also been a stimulus to explore the frontiers of non-equilibrium thermodynamics, going beyond local equilibrium approximations. Although in many situations non-equilibrium contributions to local entropy are small, in some circumstances they may be relevant, especially from a conceptual point of view [23,24]. A closely related frontier in miniaturization is the thermodynamics of small systems [30,32,39], which are also related to the Knudsen number, because what makes a system small is not its size but the number of particles it contains, and the relation between the rate of its internal collisions and collisions with the walls, or between the relaxation time and the characteristic rate of energy transfer with the outside. Systems as small as atomic nuclei have been considered as hydrodynamic and thermodynamic systems, at least around equilibrium, because they are so dense that the mean free path is smaller than the size of the system.

However, in relativistic nuclear collisions, this assumption breaks down, because the energy transferred by the fast collision to the nuclei may be comparable to the average energy of each nucleus, such that nucleons inside the nuclei do not have enough time to equilibrate their energy. Nowadays, fluxes of people, goods, and capital are very intense and have become ballistic, in the sense that all of them may go from one place to another, distant place without almost no interaction with their respective counterparts along the way. The high values of these fluxes is such that, on some occasions, the rate of exchange of people, capital, or goods is much faster than the time it takes to for the respective situation to equilibrate (from a social or a political aspect). This may bring the system very far from local equilibrium, leading in some cases to sociological and cultural conflicts.

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