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## From ballistic to diffusive regimes in heat transport at nano-scales

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### ABSTRACT

Our purpose is to model heat transport at micro- and nano-scales. At these scales, the mechanism of transport is not only diffusive, i.e. dominated by multiple scattering of phonons, but includes also a ballistic contribution due to collisions of phonons with the boundaries. To take these effects into account, we follow the line of thoughts of Extended Irreversible Thermodynamics. The original idea underlying the present paper is to assume the coexistence of two kinds of heat carriers, namely ballistic and diffusive phonons, which are obeying different time-evolution equations: the diffusive collisions are described by a Cattaneo equation and ballistic interactions by a Guyer–Krumhansl like equation. The model is applied to the problem of transient heat transport in microfilms and the results are compared with earlier descriptions.

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### 1. Introduction

Micro- and nano-scale systems have known an extraordinary fast development during the last decade due to the advancement of thin film technology and nano-devices. At these small sizes, the usual laws of macroscopic continuum physics should be revisited to take into account of the boundary and interface effects as well as the finite relaxation time of energy carriers. In particular, it has been recognized that the Fourier law of heat conduction breaks down at nano-scales: it is only applicable at large wave-length and large time scales. Indeed, it was noticed, some decades ago, that Fourier's law is not applicable to high-frequency processes as it predicts propagation of signals with an infinite velocity. Recently, G. Chen [1] has proposed an interesting model, the so-called ballistic-diffusion equation, in order to describe transient heat conduction across thin films. Chen's approach, based on Boltzmann's equation, consists in decomposing the distribution function in two parts: a diffusive and a ballistic one. As a consequence, the total (specific) internal energy  $u$  will be decomposed into a diffusive  $u_d$  and a ballistic contribution  $u_b$  in such a way that  $u = u_d + u_b$ . Similarly the heat flux vector  $\mathbf{q}$  will be split into a diffusive  $\mathbf{q}_d$  and a ballistic  $\mathbf{q}_b$  contribution so that  $\mathbf{q} = \mathbf{q}_d + \mathbf{q}_b$ .

Our objective is to show that Chen's results can be described and recovered within the framework of Extended Irreversible Thermodynamics [2,3]. The present study complements Chen's approach which is essentially based on Boltzmann's equation and the kinetic theory. It allows also to enlighten some results derived by Chen and may open interesting perspectives for future developments. The paper will run as follows. The main deficiencies of the classical Fourier and Cattaneo [4] equations as well as the ingredients of Irreversible Thermodynamics are briefly recalled in Section 2. In the next section, a thermodynamic model of heat conduction at nano-scales, involving the coexistence of ballistic and diffusive heat carriers, is proposed. The description is applied in Section 4 to the study of transient heat conduction in thin microfilms; the results are compared with these provided by other descriptions. Final comments and conclusion are drawn in Section 5.

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## 2. Diffusive and ballistic heat carriers

For isotropic systems, heat conduction is well described by the classical Fourier law

$$\mathbf{q} = -\lambda \nabla T \quad (1)$$

which relates linearly the heat flux vector  $\mathbf{q}$  to the gradient of temperature  $T$ ,  $\lambda$  designating the heat conductivity. This relation implies an instantaneous response of the system and is therefore no longer valid when the perturbation is very fast or when the response time is very long. This happens, in particular, in heat conduction in miniaturized systems, such as nano-wires and thin layers submitted to high-frequency perturbations. Heat transport in such systems differs significantly from the predictions of Fourier's law, because their behavior is strongly influenced by memory and non-local effects. Moreover, at high frequencies, one has also to introduce the relaxation time of the heat flux to take into account of the finite response time to perturbations. This has motivated the introduction of a more general heat transport equation referred to as Cattaneo's equation [4]

$$\tau \dot{\mathbf{q}} + \mathbf{q} = -\lambda \nabla T \quad (2)$$

wherein  $\tau$  denotes a suitable relaxation time, while a superposed dot stands for the material time derivative which, in the case of rigid bodies at rest considered in the present work, is identified as the partial time derivative. When  $\tau$  is negligible, or when the time variation of the heat flux is slow, Eq. (2) will reduce to Fourier law. It is worth to stress that Cattaneo's equation (2) is not the most general one to describe heat conduction in miniaturized systems. For instance, Chen [1] regarded the transport of heat in nanosize devices as a combination of ballistic phonons described by Boltzmann's kinetic equation and diffusive phonons obeying Cattaneo's equation (2). In the present work, we adopt Chen's point of view assuming the coexistence of ballistic and the diffusive heat carriers. However, in contrast with Chen, we do not refer to the kinetic Boltzmann equation but assume that ballistic phonons satisfy a time-evolution equation incorporating non-local effects. Such an attitude is typical of Extended Irreversible Thermodynamics (EIT) wherein the dissipative fluxes are elevated to the status of independent variables at the same footing as the classical variables such as temperature, or energy.

## 3. State variables and time-evolution equations

In nano- and micro-systems, the ratio between the mean free path of the heat carriers (phonons) and the characteristic length of the system given by the Knudsen number becomes comparable or larger than one. The mechanism of heat transfer is no longer of diffusive nature, i.e. dominated by multiple scattering between the carriers but it includes ballistic contributions resulting essentially from collisions with the boundaries. To cope with the ballistic effects, we shall with Chen [1] decompose the specific internal energy  $u$  and the heat flux vector  $\mathbf{q}$  into a diffusive and a ballistic contributions, and select the following set of state variables:

- i) the couple  $u_d, \mathbf{q}_d$  to account for the diffusive behavior of the heat carriers;
- ii) the couple  $u_b, \mathbf{q}_b$  to provide a description of the ballistic microscopic motions.

To summarize, the space of state variables is constituted by the quantities:  $u_d, \mathbf{q}_d$ , and  $u_b, \mathbf{q}_b$ . For future use, we introduce also the diffusive and ballistic temperatures  $T_d$  and  $T_b$  defined respectively by  $T_d = u_d/C_d$ ,  $T_b = u_b/C_b$  where  $C_d$  and  $C_b$  have the dimension of a heat capacity per unit volume. Although these quantities bear some analogy with the classical definition of the temperature, it should however be realized that, strictly speaking, these quantities do not represent temperatures in the usual sense but must be considered as a measure of the internal energies.

The next step consists in establishing the evolution equations for the set of basic variables. The evolutions of the internal energies  $u_d$  and  $u_b$  are governed by balance laws of the form

$$\dot{u}_d = -\nabla \cdot \mathbf{q}_d + r_d, \quad \dot{u}_b = -\nabla \cdot \mathbf{q}_b + r_b \quad (3)$$

while the total internal energy  $u = u_d + u_b$  satisfies the first law of thermodynamics

$$\dot{u} = -\nabla \cdot \mathbf{q} + r \quad (4)$$

the quantities  $r_d, r_b$  and  $r$  designate source terms which may be either positive or negative, in virtue of the first law (4), one has to satisfy  $r_d + r_b = r$ ; in absence of energy source ( $r = 0$ ), one has simply  $r_d = -r_b$ . Based on kinetic theory considerations, it is shown that [1]

$$r_b = -u_b/\tau_b \quad (5)$$

where the sign minus indicates that ballistic carriers can be converted into diffusive ones but that the converse is not possible,  $\tau_b$  is the relaxation time of the ballistic energy flux  $\mathbf{q}_b$ .

It remains to derive the evolution equation for the fluxes. Concerning the diffusive phonons, it is assumed that they satisfy Cattaneo's equation to cope with their high-frequency properties and in agreement with the general results of EIT; as a consequence, we are allowed to write

$$\tau_d \dot{\mathbf{q}}_d + \mathbf{q}_d = -\lambda_d \nabla T_d \quad (6)$$

wherein the relaxation time  $\tau_d$  and the heat conductivity coefficient  $\lambda_d$  are positive quantities. However, expression (6) is not able to describe the ballistic regime where most of the carriers cross the system without experiencing collisions, except with the boundaries. To cope with this situation, it is necessary to introduce non-locality. The simplest way to proceed is to complement (6) with additional terms involving  $\nabla^2 \mathbf{q}$  and  $\nabla \nabla \cdot \mathbf{q}$  so that the evolution equation of the ballistic heat flux will take the form

$$\tau_b \dot{\mathbf{q}}_b + \mathbf{q}_b = -\lambda_b \nabla T + l_b^2 (\nabla^2 \mathbf{q}_b + 2 \nabla \nabla \cdot \mathbf{q}_b) \quad (7)$$

$l_b$  is related to the correlation length of the ballistic phonons which will be identified with the characteristic length  $L$  of the system. Expression (7) has been justified within the framework of EIT [2] by selecting the flux of the flux as extra variable besides  $\mathbf{q}$ . Relation (7) is comparable to Guyer and Krumhansl's equation [2,3] obtained from the phonon kinetic theory, the latter providing also a relation between heat conductivity  $\lambda$ , relaxation time  $\tau$ , heat capacity per unit volume  $C_v$  and sound velocity  $c_0$  of the form

$$\lambda = \frac{1}{3} C_v c_0^2 \tau \quad (8)$$

Expressions (3), (6) and (7) provide the basic set of eight scalar evolution equations for the eight unknowns  $u_d$ ,  $u_b$ ,  $\mathbf{q}_d$  and  $\mathbf{q}_b$ . After eliminating  $\mathbf{q}_d$  and  $\mathbf{q}_b$  and assuming that all the transport coefficients are constant, one is led to the two second-order linear coupled differential equations:

$$\tau_d \ddot{u}_d + \dot{u}_d - (\tau_d / \tau_b) \dot{u}_b - (1 / \tau_b) u_b = (\lambda_d / C_d) \nabla^2 u_d \quad (9)$$

$$\tau_b \ddot{u}_b + 2 \dot{u}_b + u_b / \tau_b = (\lambda_b / C_b) \nabla^2 u_b + 3 l_b^2 \nabla \cdot [\nabla \dot{u}_b + (1 / \tau_b) \nabla u_b] \quad (10)$$

Expressions (9) and (10) are the key relations of our model. In the particular case  $\tau_d = \tau_b = \tau$ , making use of (3b) and setting  $l_b = 0$ , one recovers from expression (9) Chen's result

$$\tau \ddot{u}_d + \dot{u}_d + \nabla \cdot \mathbf{q}_b = (\lambda_d / C_d) \nabla^2 u_d \quad (11)$$

which differs from the telegraph equation that should be obtained from Cattaneo's law by the presence of the term in  $\nabla \cdot \mathbf{q}_b$ . In Chen's formalism, the heat flux vector  $\mathbf{q}_b$  has been obtained by using the kinetic definition of the heat flux and by solving Boltzmann's equation.

Here, we will not refer to a kinetic approach and solve the problem exclusively at the macroscopic level. As illustration, we apply the above considerations to the study of transient heat transport through a thin film whose two faces are subject to a temperature difference. Numerical checking indicates that the results of our model (Eqs. (9) and (10)) are comparable to these of Chen.

It should however be underlined that our model is more general than Chen's who introduced, without any justification, the simplifying assumptions that  $\tau_d = \tau_b$  and  $C_d = C_b$ . Chen's model is the result of a mixing of macroscopic and microscopic approaches while our description is purely macroscopic. Finally, it should be noticed that Chen remains silent about the signs of  $\tau_i$ ,  $\lambda_i$  and  $C_i$  ( $i = d, b$ ).

#### 4. Application: transient heat transport in thin films

The foregoing model will be applied to the study of heat conduction in a one-dimensional thin film of thickness  $L$  smaller than the mean free path  $l$  of the phonons, heat capacity and heat conductivity are assumed to be constant and to take the same values for the diffusive and ballistic phonons, internal energy sources are absent ( $r = 0$ ). Initially, the system is at uniform temperature  $T_0$ . The lower surface  $x = 0$  is suddenly brought to temperature  $T_1 = T_0 + \Delta T$ , while the upper surface  $x = L$  is kept at temperature  $T_0$ . Since the ballistic phonons originate at the walls, their energy is the prescribed energy at the boundaries, i.e.  $C_v T_0$  and  $C_v T_1$ , respectively. For further purpose, let us introduce the Knudsen number  $Kn = l/L$ ; since the mean free path of the ballistic carriers is comparable to the length of the material, one has  $Kn_b \approx 1$ , while for the diffusive phonons, it will be given the more general form  $(Kn_d)^2 = 3 \lambda \tau_d / C_v L^2$  in virtue of expression (8) and the result  $c_0 = l_d / \tau_d$ .

Having in mind numerical solutions, it is convenient to use dimensionless quantities

$$t^* = t / \tau_b, \quad x^* = x / L, \quad \theta_{b,d} = (u_{b,d} - C_v T_0) / C_v \Delta T, \quad \theta = (u - 2 C_v T_0) / C_v \Delta T \quad (12)$$

with  $\theta_b$ ,  $\theta_d$  and  $\theta$  ( $= \theta_b + \theta_d$ ) designating the non-dimensional temperature associated to the ballistic, diffusive and total energies respectively. The corresponding evolution equations (9) and (10) take now the form

$$\frac{\tau_d}{\tau_b} \frac{\partial^2 \theta_d}{\partial t^{*2}} - \frac{(Kn_b)^2}{3} \frac{\partial^2 \theta_d}{\partial x^{*2}} + \frac{\partial \theta_d}{\partial t^*} - \frac{\tau_d}{\tau_b} \frac{\partial \theta_b}{\partial t^*} - \theta_b - \frac{T_0}{\Delta T} = 0 \tag{13}$$

$$\frac{\partial^2 \theta_b}{\partial t^{*2}} + 2 \frac{\partial \theta_b}{\partial t^*} + \theta_b + \frac{T_0}{\Delta T} - \frac{10}{3} (Kn_b)^2 \frac{\partial^2 \theta_b}{\partial x^{*2}} - 3 (Kn_b)^2 \frac{\partial^2}{\partial x^{*2}} \frac{\partial \theta_b}{\partial t^*} = 0 \tag{14}$$

It remains to fix the initial and boundary conditions. Initially, the sample is at uniform temperature  $T_0$  (or  $u(x, 0) = C_v T_0$ ) from which follows that the corresponding non-dimensional total energy is

$$\theta_b(x^*, 0) + \theta_d(x^*, 0) = -T_0/\Delta T \tag{15}$$

As it is reasonable to admit that at short times, ballistic collisions are dominant so that the whole initial energy is of ballistic nature with  $u_b(x, 0) = C_v T_0$  and, in addition, assuming that the time derivatives of both ballistic and diffusive energies are vanishing, we are allowed to write

$$t = 0: \theta_b(x^*, 0) = 0, \quad \theta_d(x^*, 0) = -T_0/\Delta T \tag{16}$$

$$\partial \theta_b / \partial t^* = \partial \theta_d / \partial t^* = 0 \tag{17}$$

As boundary conditions, we select

$$x^* = 0: \theta_b(0, t^*) = 1, \quad \partial \theta_d / \partial x^* = 0$$

$$x^* = 1: \theta_b(1, t^*) = 0, \quad \partial \theta_d / \partial x^* = 0 \tag{18}$$

this choice is motivated by the fact that ballistic collisions originates principally at the boundaries and that the boundaries are supposed to be impervious to diffusive phonons. Preliminary numerical analyses indicate that the temperature profile is systematically lower than with Fourier's model but a good agreement with Chen's and kinetic theory results is achieved.

**5. Conclusions**

A thermodynamic description of transient heat transport at nano-scales based on Extended Irreversible Thermodynamics is proposed. The central assumption of the present work is that, contrary to previous approaches, the set of variables – namely the internal energy and the energy flux – is split into contributions of diffusive and ballistic nature. *Heat transport is viewed as a two-fluid diffusion–reaction process with ballistic particles converting into diffusive ones.*

The corresponding time-evolution equations for the ensemble of variables have been formulated with a special status reserved to ballistic carriers whose behavior is mainly dictated by non-local effects. After elimination of the heat fluxes, one has obtained two second-order coupled differential equations for the ballistic and diffusive internal energies which constitute the basic relations of our model. Our thermodynamic phenomenological approach is inspired by a recent analysis [1] by Chen. This author has developed a mixed macro-microscopic formalism wherein the diffusive regime is governed by Cattaneo's equation while ballistic particles are described by Boltzmann's equation. Chen's approach was recently shown to be in good agreement with pure kinetic results. We are not only able to reproduce the main results of Chen's ballistic-diffusion model but we have taken advantage of the thermodynamic approach to better apprehend Chen's theory.

One of our objectives was to convince the reader of the flexibility and wide range of applicability of Extended Irreversible Thermodynamics. It is shown that a rather simple model is able to cope with much of the results derived from more sophisticated approaches.

It should nevertheless be realized that the formalism discussed above represents only a first step towards a more elaborate description of heat transport at micro- and nano-scales. In particular, it must be noted that the selection of state variables is not unique. Another choice, like the total and ballistic energies together with their corresponding heat fluxes, is also allowed and would yield results similar to these of the present work. It is also expected that higher-order fluxes (the flux of the fluxes, the flux of the flux of the fluxes, etc.) [5] should be introduced from the start to cope the particulate behavior of heat carriers at short wave lengths, but the difficulty is then the physical interpretation of the new variables coupled to the complexity of the mathematical formalism. The selection of the most appropriate set of state variables remains an open problem. In addition, for the sake of simplicity, all coupling terms have been omitted in the Cattaneo and Guyer–Krumhansl relations (6) and (7), a more refined approach should include such contributions. Moreover, the establishment of appropriate boundary conditions remains a delicate task. From a more fundamental point of view, questions may also be raised about the definition of temperature at nano-scales. To circumvent this problem in our analysis, temperature was understood as a measure of internal energy.

All these considerations lead us naturally to the conclusion that heat transport in nano- and micro-systems remains a challenging, unfinished task.

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