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GENERALIZED HEAT TRANSPORT EQUATIONS IN THREE-DIMENSIONAL ANISOTROPIC RIGID HEAT CONDUCTORS*

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Abstract

In this paper we derive generalized heat transport laws for an anisotropic rigid heat conductor. We use a model formulated in a previous paper in the framework of non-equilibrium thermodynamics with internal variables. In the thermodynamic state vector beside the internal energy and the heat flux a second order tensor is introduced as internal variable, influencing the thermal phenomena. In the three-dimensional case the phenomenological equations are presented, the entropy production is worked out and anisotropic transport equations for the heat flux are carried out, as Maxwell-Cattaneo and Guyer-Krumhansl equations, describing heat waves and the ballistic propagation of the phonons, respectively. The conductivity matrix is given in the Appendix. The obtained results have applications in several technological sectors, as in nanotechnology, where there are situations of high-frequency waves propagation and Knudsen number is comparable or larger than unity.
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1 Introduction

In a previous paper [1] generalized ballistic-conductive transport heat equations were derived for isotropic rigid heat conductors, in the framework of non-equilibrium thermodynamics with internal variables (see [2], [3], [4], [5], [6], [7], [8], [9]). Generalizations of Fourier law were also given in [10], [11], [12], [13], [14], [15], [16], [17], [18], [19]. The introduction of internal variables as independent variables in the thermodynamic state vector permits to describe the internal structure of materials (see for instance [6], [20], [21], [23], [24], [25], [26], [27]). The obtained rate equations for the internal variables are different than constitutive equations and, added to the balance equations, permit to close the system of equations describing the medium under consideration. The internal variables need suitable methods to be measured [8]. In this article, using the formulation developed in [1], we focus our attention on a three-dimensional anisotropic rigid heat conductor and we derive generalized heat transport laws, having applications in several fields, as in nanotechnology. In the nanostructures the volume element size L along a direction is comparable or smaller than the free mean path l of the heat carriers (the phonons), i.e. the Knudsen number $\frac{l}{L} \geq 1$, and the rate variation of properties of these media are faster than the time scale of the relaxation times of the fluxes to their equilibrium values.

The paper is organized as follows. In Section 2 the model developed in [1] is introduced. The thermodynamic state vector is chosen, where beside the internal energy and the heat flux a second order tensor is introduced as internal variable, influencing the thermal phenomena, and assumed as odd function of microscopic particles velocities of the medium. Furthermore, in the three-dimensional and anisotropic case the phenomenological equations are presented. In Sections 3 and 4 Onsager-Casimir relations are written and the form of the entropy production is worked out. A detailed form of the conductivity matrix is given in the Appendix. In Sections 5, 6 and 7 some generalizations of the classical Fourier law, describing thermal signals having infinite velocity and relaxation time null, are carried out. In particular, the anisotropic Maxwell-Cattaneo and Guyer-Krumhansl equations, describing second-sound phenomena (heat waves) and the ballistic motion of the phonons, respectively, are derived (see [28]). In [28] a ballistic-diffusive model was developed assuming the coesistence of two types of phonons: the diffusive phonons, subjected to collisions with the core of the medium under consideration, and the ballistic phonons, originating at the boundaries and subjected to collisions with the walls of the medium. Ballistic phonons can be converted in diffusive carriers but it is not possible the inverse phenomenon. This ballistic-diffusion model was initially introduced by Chen in [30] and [31], where a integro-differential model is formulated, by resorting to the kinetic theory and macroscopic considerations. The distribution function is split in two contributions, one for ballistic phonons and the other for diffusive phonons. In this regard, see also [29] and [32].

2 Basic equations

In this Section we recall the model for a rigid heat conductor developed in a previous paper [1], in the framework of non-equilibrium thermodynamics with internal variables. The standard Cartesian tensor notation in a rectangular coordinate system is used and the equations governing the behavior of such medium are considered in a current configuration K_t. Thus, we assume that the thermodynamic state vector is the following

$$C = C(e, \boldsymbol{q}, \boldsymbol{Q}),$$

where e is the specific internal energy, $\boldsymbol{q}$ is the heat flux (the current density of the internal energy) and $\boldsymbol{Q}$ is a second order tensorial internal variable. It is assumed that $\boldsymbol{Q}$ contributes to the entropy and its flux. $\boldsymbol{Q}$ may be interpreted as the gradient of the heat flux (see [10], [11]), as the flux of the heat flux, and other quantities. Its physical meaning here is open. In this paper $\boldsymbol{Q}$ is chosen as odd function of microscopic particles velocities of the medium. In [1] also the case where $\boldsymbol{Q}$ is even function is treated.

We consider the following balance equations

$$\rho \dot{e} + \nabla \cdot \boldsymbol{q} = 0, \tag{1}$$

$$\rho \dot{s} + \nabla \cdot \boldsymbol{J} = \sigma^{(s)}, \tag{2}$$

where s is the specific entropy, $\boldsymbol{J}$ is the entropy flux, $\sigma^{(s)}$ is the entropy production per unit volume and per unit time, and an upper dot denotes the substantial derivative, i.e. $\dot{e} = \partial_t e + v^i e_{,i}$, where ∂_t is the partial time derivative, a comma in lower indices indicates the spatial derivation and Einstein summation convention for dummy indices is used. For the second law of thermodynamics $\sigma^{(s)} \geq 0$. We assume that the entropy flux is zero if $\boldsymbol{q} = \boldsymbol{0}$ and $\boldsymbol{Q} = \boldsymbol{0}$, therefore

$$J_i = b_{ij} q_j + B_{ijk} Q_{jk}, \tag{3}$$

where b_{ij} and B_{ijk} are tensorial constitutive functions of second and third order, respectively. They are Nyíri's multipliers [1]. Expanding the entropy function $s(e, \boldsymbol{q}, \boldsymbol{Q})$ up to second order approximation around an equilibrium state, we obtain

$$s(e, \boldsymbol{q}, \boldsymbol{Q}) = s^{(eq)}(e) - \frac{1}{2\rho} m_{ij} q_i q_j - \frac{1}{2\rho} M_{ijkl} Q_{ij} Q_{kl}, \tag{4}$$

where $s^{(eq)}(e)$ is the equilibrium entropy function depending on the internal energy and m_{ij} and M_{ijkl} are material coefficients. Thermodynamic stability requires that these tensors are positive definite. We assume that they are constant. From (4) we have the following symmetries

$$m_{ij} = m_{ji}, \quad M_{ijkl} = M_{klij}.$$

Furthermore, by virtue of (4) we introduce the following definitions

$$\frac{1}{T} = \frac{\partial s}{\partial e}, \tag{5}$$

$$\frac{\partial s}{\partial q_i} = -m_{ij} q_j, \tag{6}$$

$$\frac{\partial s}{\partial Q_{ij}} = -M_{ijkl} Q_{kl}. \tag{7}$$

Then, from (2) and (3), we have (see [1])

$$\rho \dot{s} + J_{i,i} = \sigma^{(s)}, \tag{8}$$

where $\sigma^{(s)}$, given by

$$\begin{aligned}
\sigma^{(s)} = {} & \rho \frac{ds^{(eq)}}{de} \dot{e} - \frac{1}{2} m_{ij} \dot{q}_i q_j - \frac{1}{2} m_{ij} q_i \dot{q}_j - \frac{1}{2} M_{ijkl} \dot{Q}_{ij} Q_{kl} \\
& - \frac{1}{2} M_{ijkl} Q_{ij} \dot{Q}_{kl} + b_{ij,i} q_j + b_{ij} q_{j,i} + B_{ijk,i} Q_{jk} + B_{ijk} Q_{jk,i} = \\
= {} & \left(b_{ij} - \frac{1}{T} \delta_{ij} \right) q_{j,i} + \left(b_{ji,j} - m_{ij} \dot{q}_j \right) q_i \\
& + \left(B_{kij,k} - M_{ijkl} \dot{Q}_{kl} \right) Q_{ij} + B_{ijk} Q_{jk,i} \geq 0,
\end{aligned} \tag{9}$$

is a non-negative definite bilinear form in the following fields: the heat flux q_i, its gradient $q_{i,j}$, the internal tensorial variable Q_{ij} and its gradient $Q_{ij,k}$. The inequality (9) expresses the second law of thermodynamics. Following the procedures of non-equilibrium thermodynamics (see [1]) we choose the fields $\left(b_{ij} - \frac{1}{T}\delta_{ij}\right)$, $(b_{ji,j} - m_{ij}\dot{q}_j)$, $\left(B_{kij,k} - M_{ijkl}\dot{Q}_{kl}\right)$ and B_{ijk} as thermodynamic fluxes and the fields q_i, $q_{i,j}$, Q_{ij} and $Q_{ij,k}$ as thermodynamic forces, so that we obtain the following *anisotropic linear phenomenological equations* between the thermodynamic fluxes and forces, where we have taken into account the physical dimensions of the present physical quantities

$$b_{ji,j} - m_{ij}\dot{q}_j = L^{(1,1)}_{ij} q_j + L^{(1,2)}_{ijk} q_{j,k} + L^{(1,3)}_{ijk} Q_{jk} + L^{(1,4)}_{ijkl} Q_{jk,l} \tag{10}$$

$$b_{ij} - \frac{1}{T}\delta_{ij} = L^{(2,1)}_{ijk} q_k + L^{(2,2)}_{ijkl} q_{k,l} + L^{(2,3)}_{ijkl} Q_{kl} + L^{(2,4)}_{ijklm} Q_{kl,m} \tag{11}$$

$$B_{kij,k} - M_{ijkl}\dot{Q}_{kl} = L^{(3,1)}_{ijk} q_k + L^{(3,2)}_{ijkl} q_{k,l} + L^{(3,3)}_{ijkl} Q_{kl} + L^{(3,4)}_{ijklm} Q_{kl,m} \tag{12}$$

$$B_{ijk} = L^{(4,1)}_{ijkl} q_l + L^{(4,2)}_{ijklm} q_{l,m} + L^{(4,3)}_{ijklm} Q_{lm} + L^{(4,4)}_{ijklmn} Q_{lm,n}. \tag{13}$$

The tensors $L^{(1,1)}_{ij}$, $L^{(1,2)}_{ijk}$, $L^{(1,3)}_{ijk}$ etc. which occur in (10)-(13) are called phenomenological tensors. They are assumed constant. For instance, $L^{(1,1)}_{ij}$ is the second order thermal conductivity tensor, connected with the influence of the heat flux on the field $(b_{ji,j} - m_{ij}\dot{q}_j)$, $L^{(1,3)}_{ijk}$ is a third order tensor connected with the influence of the internal variable Q_{ij} on the same field.

3 Onsager-Casimir relations

From phenomenological equations (10)-(13) Onsager-Casimir relations can be derived (see [1]). We choose the field Q_{ij}, and then also the field $Q_{ij,k}$, *odd* functions of the microscopic particles velocities of the medium. Furthermore, also the fields q_i, $q_{i,j}$ are *odd*, while $(b_{ji,j} - m_{ij}\dot{q}_j)$, $\left(b_{ij} - \frac{1}{T}\delta_{ij}\right)$, $\left(B_{kij,k} - M_{ijkl}\dot{Q}_{kl}\right)$ and B_{ijk} are *even* functions of these velocities (because conjugated to the odd functions, see (9)). Hence, according to the usual procedure of non-equilibrium thermodynamics, we have the following Onsager-Casimir reciprocity relations [6]

$$L^{(1,1)}_{ik} = L^{(1,1)}_{ki}, \qquad L^{(1,2)}_{ijk} = L^{(2,1)}_{jki}, \tag{14}$$

$$L^{(1,3)}_{ijk} = L^{(3,1)}_{jki}, \qquad L^{(1,4)}_{ijkl} = L^{(4,1)}_{jkli}, \tag{15}$$

$$L^{(2,2)}_{ijkl} = L^{(2,2)}_{klij}, \qquad L^{(2,3)}_{ijkl} = L^{(3,2)}_{klij}, \tag{16}$$

$$L^{(2,4)}_{ijklm} = L^{(4,2)}_{klmij}, \qquad L^{(3,3)}_{ijkl} = L^{(3,3)}_{klij}, \tag{17}$$

$$L^{(3,4)}_{ijklm} = L^{(4,3)}_{klmij}, \qquad L^{(4,4)}_{ijklmn} = L^{(4,4)}_{lmnijk}. \tag{18}$$

4 Entropy production

In this paper with the aid of (10)-(13) we derive the entropy production (9) in the three-dimensional anisotropic materials in the following form

$$
\begin{aligned}
\sigma^{(s)} = {} & L^{(1,1)}_{ik} q_i q_k + \left(L^{(1,2)}_{kji} + L^{(2,1)}_{ijk} \right) q_k q_{j,i} + \left(L^{(1,3)}_{kij} + L^{(3,1)}_{ijk} \right) q_k Q_{ij} \\
& + \left(L^{(1,4)}_{ijkl} + L^{(4,1)}_{ljki} \right) q_i Q_{jk,l} + L^{(2,2)}_{ijkl} q_{j,i} q_{k,l} + \left(L^{(2,3)}_{ijkl} + L^{(3,2)}_{klji} \right) q_{j,i} Q_{kl} \\
& + \left(L^{(2,4)}_{ijklm} + L^{(4,2)}_{mklji} \right) q_{j,i} Q_{kl,m} + L^{(3,3)}_{ijkl} Q_{ij} Q_{kl} \\
& + \left(L^{(3,4)}_{ijklm} + L^{(4,3)}_{mklij} \right) Q_{ij} Q_{kl,m} + L^{(4,4)}_{ijklmn} Q_{jk,i} Q_{lm,n} \geq 0. \qquad (19)
\end{aligned}
$$

The inequality (19) may assume the form:

$$
\begin{aligned}
\sigma^{(s)} = {} & L^{(1,1)}_{ik} q_i q_k + L^{(1,2)}_{ikl} q_i q_{k,l} + L^{(1,3)}_{ikl} q_i Q_{kl} + L^{(1,4)}_{ilmn} q_i Q_{lm,n} \\
& + L^{(2,1)}_{jik} q_{i,j} q_k + L^{(2,2)}_{jikl} q_{i,j} q_{k,l} + L^{(2,3)}_{jikl} q_{i,j} Q_{kl} + L^{(2,4)}_{jilmn} q_{i,j} Q_{lm,n} \\
& + L^{(3,1)}_{ijk} Q_{ij} q_k + L^{(3,2)}_{ijkl} Q_{ij} q_{k,l} + L^{(3,3)}_{ijkl} Q_{ij} Q_{kl} + L^{(3,4)}_{ijlmn} Q_{ij} Q_{lm,n} \\
& + L^{(4,1)}_{pijk} Q_{ij,p} q_k + L^{(4,2)}_{pijkl} Q_{ij,p} q_{k,l} + L^{(4,3)}_{pijkl} Q_{ij,p} Q_{kl} + L^{(4,4)}_{pijlmn} Q_{ij,p} Q_{lm,n} \\
& \geq 0. \qquad (20)
\end{aligned}
$$

We can also use Onsager-Casimir relations (14)-(18) and, appropriately renaming the dummy indices, entropy production (19) reads

$$
\begin{aligned}
\sigma^{(s)} = {} & L^{(1,1)}_{ik} q_i q_k + \left(L^{(1,2)}_{ikl} + L^{(1,2)}_{ilk} \right) q_i q_{k,l} + 2 L^{(1,3)}_{ikl} q_i Q_{kl} \\
& + \left(L^{(1,4)}_{ilmn} + L^{(1,4)}_{inlm} \right) q_i Q_{lm,n} + L^{(2,2)}_{jikl} q_{i,j} q_{k,l} + \left(L^{(2,3)}_{jikl} + L^{(2,3)}_{ijkl} \right) q_{i,j} Q_{kl} \\
& + \left(L^{(2,4)}_{jilmn} + L^{(2,4)}_{ijnlm} \right) q_{i,j} Q_{lm,n} + L^{(3,3)}_{ijkl} Q_{ij} Q_{kl} \\
& + \left(L^{(3,4)}_{ijlmn} + L^{(3,4)}_{ijnlm} \right) Q_{ij} Q_{lm,n} + L^{(4,4)}_{pijlmn} Q_{ij,p} Q_{lm,n} \geq 0. \qquad (21)
\end{aligned}
$$

From (20) it is seen that the entropy production in a non-negative bilinear form in the components of the heat flux, the components of the spatial derivatives of the heat flux, the components of the internal variable and the components of the spatial derivatives of the internal variable (see in Appendix its matrix representation). Therefore, several inequalities can be obtained for the components of the phenomenological tensors, resulting from the fact that all the elements of the main diagonal of the symbolic matrix

$\{L_{\alpha\beta}\}$ associated to the bilinear form (20) must be non-negative (condition only necessary for the semi-definiteness of the matrix $\{L_{\alpha\beta}\}$), i.e.

$$L_{ii}^{(1,1)} \geq 0, \quad L_{jiij}^{(2,2)} \geq 0, \quad L_{ijij}^{(3,3)} \geq 0, \quad L_{pijijp}^{(4,4)} \geq 0, \tag{22}$$

for all $i, j, p = 1, 2, 3$, (in particular $L_{iiii}^{(2,2)} \geq 0$, $L_{iiii}^{(3,3)} \geq 0$, $L_{iiiiii}^{(4,4)} \geq 0$ for all $i = 1, 2, 3$). Furthermore, from Sylverster's criterion (necessary and sufficient condition for the semi-definiteness of the matrix $\{L_{\alpha\beta}\}$) all the principal minors P_r of $\{L_{\alpha\beta}\}$, defined by

$$P_r = \begin{vmatrix} p_{11} & p_{12} & \dots & p_{1r} \\ p_{21} & p_{22} & \dots & p_{2r} \\ \dots & \dots & \dots & \dots \\ p_{r1} & p_{r2} & \dots & p_{rr} \end{vmatrix} \tag{23}$$

for all $r = 1, \dots, 48$, of even and odd order, must also be non-negative.
For example, from the non-negativity of the first two principal minors, we have

$$L_{11}^{(1,1)} \geq 0, \qquad L_{11}^{(1,1)} L_{22}^{(1,1)} - \left(L_{12}^{(1,1)}\right)^2 \geq 0. \tag{24}$$

5 A generalized anisotropic heat transport equation

From equations (11) and (13) we have:

$$b_{ji,j} = \left(\frac{1}{T}\right)_{,i} + L_{jik}^{(2,1)} q_{k,j} + L_{jikl}^{(2,2)} q_{k,lj} + L_{jikl}^{(2,3)} Q_{kl,j} + L_{jiklm}^{(2,4)} Q_{kl,mj} \tag{25}$$

$$B_{kij,k} = L_{kijl}^{(4,1)} q_{l,k} + L_{kijlm}^{(4,2)} q_{l,mk} + L_{kijlm}^{(4,3)} Q_{lm,k} + L_{kijlmn}^{(4,4)} Q_{lm,nk}. \tag{26}$$

Using Onsager-Casimir relations (14)-(18) and equations (25), (26), we can write (10) and (12) as follows

$$\begin{aligned} m_{ij}\dot{q}_j + L_{ij}^{(1,1)} q_j + (L_{ijk}^{(1,2)} - L_{jki}^{(1,2)}) q_{j,k} - L_{jikl}^{(2,2)} q_{k,lj} = \left(\frac{1}{T}\right)_{,i} - L_{ijk}^{(1,3)} Q_{jk} \\ + (L_{lijk}^{(2,3)} - L_{ijkl}^{(1,4)}) Q_{jk,l} + L_{jiklm}^{(2,4)} Q_{kl,mj}, \end{aligned} \tag{27}$$

$$\begin{aligned} & M_{ijkl}\dot{Q}_{kl} + L_{ijkl}^{(3,3)} Q_{kl} + (L_{ijlmk}^{(3,4)} - L_{lmkij}^{(3,4)}) Q_{lm,k} - L_{kijlmn}^{(4,4)} Q_{lm,nk} \\ & = (L_{lkij}^{(1,4)} - L_{lkij}^{(2,3)}) q_{l,k} - L_{kij}^{(1,3)} q_k + L_{lmkij}^{(2,4)} q_{l,mk}. \end{aligned} \tag{28}$$

Assuming that $L^{(3,4)}_{ijlmk}$, M_{ijkl} and $L^{(4,4)}_{kijlmn}$ are negligible, equation (28) takes the form

$$L^{(3,3)}_{ijkl} Q_{kl} = (L^{(1,4)}_{lkij} - L^{(2,3)}_{lkij}) q_{l,k} - L^{(1,3)}_{kij} q_k + L^{(2,4)}_{lmkij} q_{l,mk}. \tag{29}$$

We assume that it is possible to define the inverse of $L^{(3,3)}_{ijkl}$, $(L^{(3,3)}_{hpji})^{-1}$, such that

$$(L^{(3,3)}_{hpji})^{-1} L^{(3,3)}_{ijkl} = L^{(3,3)}_{hpji} (L^{(3,3)}_{ijkl})^{-1} = \delta_{hl}\delta_{pk}. \tag{30}$$

Recalling that if H_{ijkl} and M_{ijkl} are two fourth order tensors, the double inner product between them is defined by $J_{ijkl} \coloneqq H_{ijhp} M_{phkl}$, the inverse of a fourth order tensor L_{ijkl} is defined by $(L_{ijhp})^{-1} L_{phkl} = L_{ijhp} (L_{phkl})^{-1} = \delta_{il}\delta_{jk}$, with $\delta_{il}\delta_{jk} = I_{ijkl}$, I_{ijkl} being the fourth order identity tensor (see [33]). Multiplying both sides of (29) by $(L^{(3,3)}_{hpji})^{-1}$ and summing over i and j, we derive the following expression for the internal variable $\boldsymbol{Q}$

$$Q_{ph} = (L^{(3,3)}_{hpji})^{-1} (L^{(1,4)}_{lkij} - L^{(2,3)}_{lkij}) q_{l,k} - (L^{(3,3)}_{hpji})^{-1} L^{(1,3)}_{kij} q_k + (L^{(3,3)}_{hpji})^{-1} L^{(2,4)}_{lmkij} q_{l,mk}. \tag{31}$$

We derive (31) with respect to x_r and x_s, so that we deduce

$$\begin{aligned} Q_{ph,r} &= (L^{(3,3)}_{hpji})^{-1} (L^{(1,4)}_{lkij} - L^{(2,3)}_{lkij}) q_{l,kr} - (L^{(3,3)}_{hpji})^{-1} L^{(1,3)}_{kij} q_{k,r} \\ &\quad + (L^{(3,3)}_{hpji})^{-1} L^{(2,4)}_{lmkij} q_{l,mkr}, \end{aligned} \tag{32}$$

$$\begin{aligned} Q_{ph,rs} &= (L^{(3,3)}_{hpji})^{-1} (L^{(1,4)}_{lkij} - L^{(2,3)}_{lkij}) q_{l,krs} - (L^{(3,3)}_{hpji})^{-1} L^{(1,3)}_{kij} q_{k,rs} \\ &\quad + (L^{(3,3)}_{hpji})^{-1} L^{(2,4)}_{lmkij} q_{l,mkrs}. \end{aligned} \tag{33}$$

Rewriting equation (27) as follows

$$\begin{aligned} m_{ij}\dot{q}_j + L^{(1,1)}_{ij} q_j + (L^{(1,2)}_{ijk} - L^{(1,2)}_{jki}) q_{j,k} - L^{(2,2)}_{jikl} q_{k,lj} = \left(\frac{1}{T}\right)_{,i} - L^{(1,3)}_{iph} Q_{ph} \\ + (L^{(2,3)}_{riph} - L^{(1,4)}_{iphr}) Q_{ph,r} + L^{(2,4)}_{siphr} Q_{ph,rs} \end{aligned} \tag{34}$$

and using (31), (32) and (33), equation (34) takes the form

$$
\begin{aligned}
& m_{ik}\dot{q}_k + \left[L^{(1,1)}_{ik} - L^{(1,3)}_{iph}\left(L^{(3,3)}_{hpjm}\right)^{-1} L^{(1,3)}_{kmj}\right] q_k + \left[L^{(1,2)}_{ilk} + L^{(1,3)}_{iph}\left(L^{(3,3)}_{hpjm}\right)^{-1}\right. \\
& \left.\left(L^{(1,4)}_{lkmj} - L^{(2,3)}_{lkmj}\right) - L^{(1,4)}_{iphk}\left(L^{(3,3)}_{hpjm}\right)^{-1} L^{(1,3)}_{lmj} - L^{(1,2)}_{lki} + L^{(1,3)}_{lmj}\left(L^{(3,3)}_{hpjm}\right)^{-1} L^{(2,3)}_{kiph}\right] q_{l,k} \\
& + \left[L^{(1,3)}_{iph}\left(L^{(3,3)}_{hpjn}\right)^{-1} L^{(2,4)}_{lrknj} + L^{(2,4)}_{kiphr}\left(L^{(3,3)}_{hpjm}\right)^{-1} L^{(1,3)}_{lmj} - L^{(2,2)}_{rilk} - L^{(2,3)}_{riph}\left(L^{(3,3)}_{hpjm}\right)^{-1}\right. \\
& \left.\left(L^{(1,4)}_{lkmj} - L^{(2,3)}_{lkmj}\right) + L^{(1,4)}_{iphr}\left(L^{(3,3)}_{hpjm}\right)^{-1}\left(L^{(1,4)}_{lkmj} - L^{(2,3)}_{lkmj}\right)\right] q_{l,kr} \\
& + \left[L^{(1,4)}_{iphr}\left(L^{(3,3)}_{hpjn}\right)^{-1} L^{(2,4)}_{lmknj} - L^{(2,4)}_{miphr}\left(L^{(3,3)}_{hpjn}\right)^{-1}\left(L^{(1,4)}_{lknj} - L^{(2,3)}_{lknj}\right)\right. \\
& \left.- L^{(2,3)}_{riph}\left(L^{(3,3)}_{hpjn}\right)^{-1} L^{(2,4)}_{lmknj}\right] q_{l,krm} - L^{(2,4)}_{siphr}\left(L^{(3,3)}_{hpjn}\right)^{-1} L^{(2,4)}_{lmknj} q_{l,krms} = \left(\frac{1}{T}\right)_{,i},
\end{aligned}
\tag{35}
$$

i.e.

$$
\begin{aligned}
& m_{ik}\dot{q}_k + \mathcal{L}^{(1)}_{(q)ik} q_k + \left(\mathcal{A}^{(1)}_{ilk} + \mathcal{A}^{(2)}_{ikl} + \mathcal{A}^{(3)}_{lki}\right) q_{l,k} \\
& \quad + \left(\mathcal{B}^{(1)}_{ilrk} + \mathcal{B}^{(2)}_{kirl} + \mathcal{B}^{(3)}_{rilk} + \mathcal{B}^{(4)}_{irlk}\right) q_{l,kr} \\
& \qquad + \left(\mathcal{C}^{(1)}_{irlmk} + \mathcal{C}^{(2)}_{mirlk} + \mathcal{C}^{(3)}_{rilmk}\right) q_{l,krm} + \mathcal{D}_{sirlmk} q_{l,krms} = \left(\frac{1}{T}\right)_{,i},
\end{aligned}
\tag{36}
$$

where

$$\mathcal{L}^{(1)}_{(q)ik} = L^{(1,1)}_{ik} - L^{(1,3)}_{iph}\left(L^{(3,3)}_{hpjm}\right)^{-1} L^{(1,3)}_{kmj}, \tag{37}$$

$$\mathcal{A}^{(1)}_{ilk} = L^{(1,2)}_{ilk} + L^{(1,3)}_{iph}\left(L^{(3,3)}_{hpjm}\right)^{-1}\left(L^{(1,4)}_{lkmj} - L^{(2,3)}_{lkmj}\right), \tag{38}$$

$$\mathcal{A}^{(2)}_{ikl} = -L^{(1,4)}_{iphk}\left(L^{(3,3)}_{hpjm}\right)^{-1} L^{(1,3)}_{lmj}, \tag{39}$$

$$\mathcal{A}^{(3)}_{lki} = -L^{(1,2)}_{lki} + L^{(1,3)}_{lmj}\left(L^{(3,3)}_{hpjm}\right)^{-1} L^{(2,3)}_{kiph}, \tag{40}$$

$$\mathcal{B}^{(1)}_{ilrk} = L^{(1,3)}_{iph}\left(L^{(3,3)}_{hpjn}\right)^{-1} L^{(2,4)}_{lrknj}, \tag{41}$$

$$\mathcal{B}^{(2)}_{kirl} = L^{(2,4)}_{kiphr}\left(L^{(3,3)}_{hpjm}\right)^{-1} L^{(1,3)}_{lmj}, \tag{42}$$

$$\mathcal{B}^{(3)}_{rilk} = -L^{(2,2)}_{rilk} - L^{(2,3)}_{riph}\left(L^{(3,3)}_{hpjm}\right)^{-1}\left(L^{(1,4)}_{lkmj} - L^{(2,3)}_{lkmj}\right), \tag{43}$$

$$\mathcal{B}^{(4)}_{irlk} = L^{(1,4)}_{iphr}\left(L^{(3,3)}_{hpjm}\right)^{-1}\left(L^{(1,4)}_{lkmj} - L^{(2,3)}_{lkmj}\right), \tag{44}$$

$$\mathcal{C}^{(1)}_{irlmk} = L^{(1,4)}_{iphr}\left(L^{(3,3)}_{hpjn}\right)^{-1} L^{(2,4)}_{lmknj}, \tag{45}$$

$$\mathcal{C}^{(2)}_{mirlk} = -L^{(2,4)}_{miphr}\left(L^{(3,3)}_{hpjn}\right)^{-1}\left(L^{(1,4)}_{lknj} - L^{(2,3)}_{lknj}\right), \tag{46}$$

$$\mathcal{C}^{(3)}_{rilmk} = -L^{(2,3)}_{riph}\left(L^{(3,3)}_{hpjn}\right)^{-1} L^{(2,4)}_{lmknj}, \tag{47}$$

$$\mathcal{D}_{sirlmk} = -L^{(2,4)}_{siphr}\left(L^{(3,3)}_{hpjn}\right)^{-1} L^{(2,4)}_{lmknj}. \tag{48}$$

5.1 Anisotropic Guyer-Krumhansl equation

In the case where $\boldsymbol{\mathcal{A}}^{(i)} = \boldsymbol{\mathcal{C}}^{(i)} = \boldsymbol{\mathcal{D}} = \mathbf{0}$ $(i = 1, 2, 3)$ equation (36) becomes

$$m_{ik}\dot{q}_k + \mathcal{L}^{(1)}_{(q)ik} q_k + \left(\mathcal{B}^{(1)}_{ilrk} + \mathcal{B}^{(2)}_{kirl} + \mathcal{B}^{(3)}_{rilk} + \mathcal{B}^{(4)}_{irlk}\right) q_{l,kr} = \left(\frac{1}{T}\right)_{,i}. \tag{49}$$

We observe that the field $q_{l,kr}$ contains three different types of terms $q_{m,mr}$, $q_{l,mm}$ $(m, r, l = 1, 2, 3$ and $l \neq m)$ and $q_{\bar{l},\bar{k}\bar{r}}$ $(\bar{l} \neq \bar{k} \neq \bar{r})$. Furthermore, by virtue of Schwarz theorem the terms of the type $q_{\bar{l},\bar{k}\bar{r}}$ are only $q_{1,23} = q_{1,32}$, $q_{2,13} = q_{2,31}$ and $q_{3,12} = q_{3,21}$ and the terms of $q_{l,kr}$ in which $l = r$ are included in the terms of the type $q_{m,mr}$ (for example $q_{1,21} = q_{1,12}$).

Therefore, when the tensor $q_{l,kr}$ assumes the special form

$$q_{l,kr} = q_{m,mr}\delta_{lk} + q_{l,mm}\delta_{kr}, \tag{50}$$

from (49) we derive the following *anisotropic Guyer-Krumhansl equation,* describing the behaviour of the ballistic phonons colliding with the boundaries of the medium, influenced by highly non-local effects characterizing small-scale systems,

$$m_{ik}\dot{q}_k + \mathcal{L}^{(1)}_{(q)ik} q_k + \left(\mathscr{L}_{ir} + \mathcal{B}^{(3)}_{rikk}\right) q_{m,mr} + \mathscr{H}_{il}\, q_{l,mm} = \left(\frac{1}{T}\right)_{,i}, \tag{51}$$

where

$$\mathscr{L}_{ir} = \mathcal{B}^{(1)}_{ikrk} + \mathcal{B}^{(2)}_{kirk} + \mathcal{B}^{(4)}_{irkk}, \quad \mathscr{H}_{il} = \mathcal{B}^{(1)}_{ilkk} + \mathcal{B}^{(2)}_{kikl} + \mathcal{B}^{(3)}_{kilk} + \mathcal{B}^{(4)}_{iklk}. \tag{52}$$

Assuming that it is possible to define the inverse of $\boldsymbol{\mathcal{L}}^{(1)}_{(q)}$, multiplying (51) by $\left(\mathcal{L}^{(1)}_{(q)ji}\right)^{-1}$, we obtain

$$\tau_{ik}\dot{q}_k + q_i + (\mathcal{L}^{(1)}_{(q)ij})^{-1}\left[(\mathscr{L}_{jr} + \mathcal{B}^{(3)}_{rjkk}) q_{m,mr} + \mathscr{H}_{jl} q_{l,mm}\right] = -\lambda_{ik} T_{,k}, \tag{53}$$

where

$$\tau_{ik} = \left(\mathcal{L}^{(1)}_{(q)ij}\right)^{-1} m_{jk} \quad \text{and} \quad \lambda_{ik} = \frac{1}{T^2}\left(\mathcal{L}^{(1)}_{(q)ik}\right)^{-1} \tag{54}$$

are the relaxation times tensor and the thermal conducibility tensor, respectively. The thermal disturbances have finite velocity of propagation. For Guyer-Krumhansl equation in the isotropic case see [1], [28], [30], [31] and [18].

6 A generalized heat transport equation derived from (36) splitting $q_{l,k}$

Splitting the field $q_{l,k}$ in its deviatoric part $\widetilde{q}_{l,k}$ and its scalar part $\frac{1}{3}q_{m,m}\delta_{lk}$ we have:

$$q_{l,k} = \widetilde{q}_{l,k} + \frac{1}{3}q_{m,m}\delta_{lk} \tag{55}$$

so that we can write:

$$q_{l,kr} = \widetilde{q}_{l,kr} + \frac{1}{3}q_{m,mr}\delta_{lk} \tag{56}$$

By virtue of (56), we have:

$$\begin{aligned}\left(\mathcal{B}^{(1)}_{ilrk} + \mathcal{B}^{(2)}_{kirl} + \mathcal{B}^{(3)}_{rilk} + \mathcal{B}^{(4)}_{irlk}\right) q_{l,kr} &= \left(\mathcal{B}^{(1)}_{ilrk} + \mathcal{B}^{(2)}_{kirl} + \mathcal{B}^{(3)}_{rilk} + \mathcal{B}^{(4)}_{irlk}\right) \widetilde{q}_{l,kr} \\ &+ \frac{1}{3}\left(\mathcal{B}^{(1)}_{ikrk} + \mathcal{B}^{(2)}_{kirk} + \mathcal{B}^{(3)}_{rikk} + \mathcal{B}^{(4)}_{irkk}\right) q_{m,mr}\end{aligned} \tag{57}$$

and equation (36) takes the form:

$$\begin{aligned}&m_{ik}\dot{q}_k + \mathcal{L}^{(1)}_{(q)ik} q_k + \left(\mathcal{A}^{(1)}_{ilk} + \mathcal{A}^{(2)}_{ikl} + \mathcal{A}^{(3)}_{lki}\right) q_{l,k} \\ &+ \left(\mathcal{B}^{(1)}_{ilrk} + \mathcal{B}^{(2)}_{kirl} + \mathcal{B}^{(3)}_{rilk} + \mathcal{B}^{(4)}_{irlk}\right) \widetilde{q}_{l,kr} \\ &\quad + \frac{1}{3}\left(\mathcal{B}^{(1)}_{ikrk} + \mathcal{B}^{(2)}_{kirk} + \mathcal{B}^{(3)}_{rikk} + \mathcal{B}^{(4)}_{irkk}\right) q_{m,mr} \\ &+ \left(\mathcal{C}^{(1)}_{irlmk} + \mathcal{C}^{(2)}_{mirlk} + \mathcal{C}^{(3)}_{rilmk}\right) q_{l,krm} \\ &\qquad + \mathcal{D}_{sirlmk} q_{l,krms} = \left(\frac{1}{T}\right)_{,i}.\end{aligned} \tag{58}$$

In the case where in (58) $\boldsymbol{\mathcal{A}}^{(i)} = \boldsymbol{\mathcal{C}}^{(i)} = \boldsymbol{\mathcal{D}} = \mathbf{0}$ $(i = 1, 2, 3)$ and the contribution of the field $\widetilde{q}_{l,kr}$ is negligible, we obtain Guyer-Krumhansl equation in the form

$$m_{ik}\dot{q}_k + \mathcal{L}^{(1)}_{(q)ik} q_k + \frac{1}{3}\left(\mathcal{B}^{(1)}_{ikrk} + \mathcal{B}^{(2)}_{kirk} + \mathcal{B}^{(3)}_{rikk} + \mathcal{B}^{(4)}_{irkk}\right) q_{m,mr} = \left(\frac{1}{T}\right)_{,i}, \tag{59}$$

where only the contribution of the gradient of the divergence of $\boldsymbol{q}$ appears. Multiplying (59) by $\mathcal{L}^{(1)}_{(q)ji})^{-1}$, we have

$$\tau_{ik}\dot{q}_k + q_i + (\mathcal{L}^{(1)}_{(q)ij})^{-1}\left[\frac{1}{3}\left(\mathcal{B}^{(1)}_{ikrk} + \mathcal{B}^{(2)}_{kirk} + \mathcal{B}^{(3)}_{rikk} + \mathcal{B}^{(4)}_{irkk}\right) q_{m,mr}\right] = -\lambda_{ik}T_{,k}, \tag{60}$$

where the relaxation times tensor τ_{ik} and the thermal conductivity tensor λ_{ik} are given by (54).

6.1 Maxwell-Vernotte-Cattaneo equation

In the case where $\boldsymbol{\mathcal{A}}^{(i)} = \boldsymbol{\mathcal{B}}^{(j)} = \boldsymbol{\mathcal{C}}^{(i)} = \boldsymbol{\mathcal{D}} = \mathbf{0}$ $(i = 1,2,3, \;\; j = 1,2,3,4)$, equations (58) and (36) become

$$m_{ik}\dot{q}_k + \mathcal{L}^{(1)}_{(q)ik}q_k = -\frac{1}{T^2}T_{,i}. \tag{61}$$

Multiplying (61) by $(\mathcal{L}^{(1)}_{(q)ji})^{-1}$, we obtain the anisotropic Maxwell-Vernotte-Cattaneo equation in the form

$$\tau_{ik}\dot{q}_k + q_i = -\lambda_{ik}T_{,k}, \tag{62}$$

where τ_{ik} and λ_{ik} are given by (54). In this case the thermal disturbances have finite propagation velocity and their own relaxation time. Equation (61) can also be obtained from (59) when $\boldsymbol{\mathcal{B}}^{(j)} = \mathbf{0}$ $(j = 1,2,3,4)$. For the isotropic Maxwell-Vernotte-Cattaneo equation see [1], [18], [28], [29]). Equation (62) governs the motion of the diffusive phonons, that are subjected to collisons with the core of the medium.

6.2 Fourier equation

In the case where $\boldsymbol{m} = \boldsymbol{\mathcal{A}}^{(i)} = \boldsymbol{\mathcal{B}}^{(j)} = \boldsymbol{\mathcal{C}}^{(i)} = \boldsymbol{\mathcal{D}} = \mathbf{0}$ $(i = 1,2,3, \;\; j = 1,2,3,4)$, equation (58) and (36) become

$$\mathcal{L}^{(1)}_{(q)ik}q_k = -\frac{1}{T^2}T_{,i}. \tag{63}$$

Equation (63) can also be obtained from (62) when the relaxation times tensor is null and describes thermal disturbances having propagation velocity infinite. Multiplying (63) by $(\mathcal{L}^{(1)}_{(q)ji})^{-1}$ we deduce the anisotropic Fourier equation

$$q_i = -\lambda_{ij}T_{,j} \tag{64}$$

where λ_{ij} is given by $(54)_2$. Equation (64) is valid at low frequencies and large space scales.

7 Conclusions

In this article we focused our attention on anisotropic rigid heat conductors and, using a theory formulated for isotropic media [1], we derived generalized heat transport laws. One of our objectives was to put in evidence the flexibility and the large range of applicability of non-equilibrium thermodynamics with internal variables, that with standard procedures permits to obtain relevant results without resorting to more elaborated approaches. A tensorial internal variable of second order, influencing the thermal phenomena and assumed as odd function of microscopic particles velocities of the medium, was introduced in the thermodynamic state vector. Three-dimensional and anisotropic transport equations for the heat flux were obtained as generalizations of the classical Fourier law, that describes thermal signals at low frequency, having infinite velocity and relaxation time null. In particular, the anisotropic Maxwell-Cattaneo and Guyer-Krumhansl equations were carried out, describing second-sound phenomena and ballistic motion of phonons that have collisions with the walls of the medium, respectively.

Appendix

In this Appendix we give a representation of the conductivity matrix $L_{\alpha\beta}$, that may be useful in numerical simulations. Entropy production (20) can also be written in the symbolic notation

$$X_\alpha L_{\alpha\beta} X_\beta \geq 0, \tag{65}$$

where

$$\begin{aligned}
&\{X_\alpha\} = \{q_i \ ; \ q_{i,j} \ ; \ Q_{ij} \ ; \ Q_{ij,p}\} = \\
&= \{q_1 \ ; \ q_2 \ ; \ q_3 \ ; \ q_{1,1} \ ; \ q_{1,2} \ ; \ q_{1,3} \ ; \ q_{2,1} \ ; \ q_{2,2} \ ; \ q_{2,3} \ ; \ q_{3,1} \ ; \ q_{3,2} \ ; \ q_{3,3} \ ; \\
&\quad Q_{11,1} \ ; \ Q_{11,2} \ ; \ Q_{11,3} \ ; \ Q_{12,1} \ ; \ Q_{12,2} \ ; \ Q_{12,3} \ ; \ Q_{13,1} \ ; \ Q_{13,2} \ ; \ Q_{13,3} \ ; \\
&\quad Q_{21,1} \ ; \ Q_{21,2} \ ; \ Q_{21,3} \ ; \ Q_{22,1} \ ; \ Q_{22,2} \ ; \ Q_{22,3} \ ; \ Q_{23,1} \ ; \ Q_{23,2} \ ; \ Q_{23,3} \ ; \\
&\quad Q_{31,1} \ ; \ Q_{31,2} \ ; \ Q_{31,3} \ ; \ Q_{32,1} \ ; \ Q_{32,2} \ ; \ Q_{32,3} \ ; \ Q_{33,1} \ ; \ Q_{33,2} \ ; \ Q_{33,3}\} \\
&\ (\alpha = 1, \ldots, 48),
\end{aligned} \tag{66}$$

$$\{X_\beta\} = \left\{ \begin{array}{c} q_k \\ q_{k,l} \\ Q_{kl} \\ Q_{lm,n} \end{array} \right\} \quad (\beta = 1, \ldots, 48), \tag{67}$$

and for $L_{\alpha\beta}$ we introduce the following notation

$$\{L_{\alpha\beta}\} = \left(\begin{array}{c|c|c|c} \overset{3\times3}{L^{(1,1)}_{ik}} & \overset{3\times9}{L^{(1,2)}_{ikl}} & \overset{3\times9}{L^{(1,3)}_{ikl}} & \overset{3\times27}{L^{(1,4)}_{ilmn}} \\ \hline \overset{9\times3}{L^{(2,1)}_{jik}} & \overset{9\times9}{L^{(2,2)}_{jikl}} & \overset{9\times9}{L^{(2,3)}_{jikl}} & \overset{9\times27}{L^{(2,4)}_{jilmn}} \\ \hline \overset{9\times3}{L^{(3,1)}_{ijk}} & \overset{9\times9}{L^{(3,2)}_{ijkl}} & \overset{9\times9}{L^{(3,3)}_{ijkl}} & \overset{9\times27}{L^{(3,4)}_{ijlmn}} \\ \hline \overset{27\times3}{L^{(4,1)}_{pijk}} & \overset{27\times9}{L^{(4,2)}_{pijkl}} & \overset{27\times9}{L^{(4,3)}_{pijkl}} & \overset{27\times27}{L^{(4,4)}_{pijlmn}} \end{array} \right) \quad (\alpha, \beta = 1, \ldots, 48). \tag{68}$$

where, for instance, $\overset{3\times3}{L^{(1,1)}_{ik}}$ indicates the generic element of a symbolic matrix of dimension 3×3 and so on. Some symbolic sub-matrices that appear in (76) have the following form

$$L^{(1,1)}_{ik} = \begin{pmatrix} L^{(1,1)}_{11} & L^{(1,1)}_{12} & L^{(1,1)}_{13} \\ L^{(1,1)}_{12} & L^{(1,1)}_{22} & L^{(1,1)}_{23} \\ L^{(1,1)}_{13} & L^{(1,1)}_{23} & L^{(1,1)}_{33} \end{pmatrix}, \tag{69}$$

$$L^{(2,2)}_{jikl} = \begin{pmatrix} L^{(2,2)}_{1111} & L^{(2,2)}_{1112} & L^{(2,2)}_{1113} & L^{(2,2)}_{1121} & L^{(2,2)}_{1122} & L^{(2,2)}_{1123} & L^{(2,2)}_{1131} & L^{(2,2)}_{1132} & L^{(2,2)}_{1133} \\ L^{(2,2)}_{2111} & L^{(2,2)}_{2112} & L^{(2,2)}_{2113} & L^{(2,2)}_{2121} & L^{(2,2)}_{2122} & L^{(2,2)}_{2123} & L^{(2,2)}_{2131} & L^{(2,2)}_{2132} & L^{(2,2)}_{2133} \\ L^{(2,2)}_{3111} & L^{(2,2)}_{3112} & L^{(2,2)}_{3113} & L^{(2,2)}_{3121} & L^{(2,2)}_{3122} & L^{(2,2)}_{3123} & L^{(2,2)}_{3131} & L^{(2,2)}_{3132} & L^{(2,2)}_{3133} \\ L^{(2,2)}_{1211} & L^{(2,2)}_{1212} & L^{(2,2)}_{1213} & L^{(2,2)}_{1221} & L^{(2,2)}_{1222} & L^{(2,2)}_{1223} & L^{(2,2)}_{1231} & L^{(2,2)}_{1232} & L^{(2,2)}_{1233} \\ L^{(2,2)}_{2211} & L^{(2,2)}_{2212} & L^{(2,2)}_{2213} & L^{(2,2)}_{2221} & L^{(2,2)}_{2222} & L^{(2,2)}_{2223} & L^{(2,2)}_{2231} & L^{(2,2)}_{2232} & L^{(2,2)}_{2233} \\ L^{(2,2)}_{3211} & L^{(2,2)}_{3212} & L^{(2,2)}_{3213} & L^{(2,2)}_{3221} & L^{(2,2)}_{3222} & L^{(2,2)}_{3223} & L^{(2,2)}_{3231} & L^{(2,2)}_{3232} & L^{(2,2)}_{3233} \\ L^{(2,2)}_{1311} & L^{(2,2)}_{1312} & L^{(2,2)}_{1313} & L^{(2,2)}_{1321} & L^{(2,2)}_{1322} & L^{(2,2)}_{1323} & L^{(2,2)}_{1331} & L^{(2,2)}_{1332} & L^{(2,2)}_{1333} \\ L^{(2,2)}_{2311} & L^{(2,2)}_{2312} & L^{(2,2)}_{2313} & L^{(2,2)}_{2321} & L^{(2,2)}_{2322} & L^{(2,2)}_{2323} & L^{(2,2)}_{2331} & L^{(2,2)}_{2332} & L^{(2,2)}_{2333} \\ L^{(2,2)}_{3311} & L^{(2,2)}_{3312} & L^{(2,2)}_{3313} & L^{(2,2)}_{3321} & L^{(2,2)}_{3322} & L^{(2,2)}_{3323} & L^{(2,2)}_{3331} & L^{(2,2)}_{3332} & L^{(2,2)}_{3333} \end{pmatrix}, \tag{70}$$

$$L^{(2,3)}_{jikl} = \begin{pmatrix} L^{(2,3)}_{1111} & L^{(2,3)}_{1112} & L^{(2,3)}_{1113} & L^{(2,3)}_{1121} & L^{(2,3)}_{1122} & L^{(2,3)}_{1123} & L^{(2,3)}_{1131} & L^{(2,3)}_{1132} & L^{(2,3)}_{1133} \\ L^{(2,3)}_{2111} & L^{(2,3)}_{2112} & L^{(2,3)}_{2113} & L^{(2,3)}_{2121} & L^{(2,3)}_{2122} & L^{(2,3)}_{2123} & L^{(2,3)}_{2131} & L^{(2,3)}_{2132} & L^{(2,3)}_{2133} \\ L^{(2,3)}_{3111} & L^{(2,3)}_{3112} & L^{(2,3)}_{3113} & L^{(2,3)}_{3121} & L^{(2,3)}_{3122} & L^{(2,3)}_{3123} & L^{(2,3)}_{3131} & L^{(2,3)}_{3132} & L^{(2,3)}_{3133} \\ L^{(2,3)}_{1211} & L^{(2,3)}_{1212} & L^{(2,3)}_{1213} & L^{(2,3)}_{1221} & L^{(2,3)}_{1222} & L^{(2,3)}_{1223} & L^{(2,3)}_{1231} & L^{(2,3)}_{1232} & L^{(2,3)}_{1233} \\ L^{(2,3)}_{2211} & L^{(2,3)}_{2212} & L^{(2,3)}_{2213} & L^{(2,3)}_{2221} & L^{(2,3)}_{2222} & L^{(2,3)}_{2223} & L^{(2,3)}_{2231} & L^{(2,3)}_{2232} & L^{(2,3)}_{2233} \\ L^{(2,3)}_{3211} & L^{(2,3)}_{3212} & L^{(2,3)}_{3213} & L^{(2,3)}_{3221} & L^{(2,3)}_{3222} & L^{(2,3)}_{3223} & L^{(2,3)}_{3231} & L^{(2,3)}_{3232} & L^{(2,3)}_{3233} \\ L^{(2,3)}_{1311} & L^{(2,3)}_{1312} & L^{(2,3)}_{1313} & L^{(2,3)}_{1321} & L^{(2,3)}_{1322} & L^{(2,3)}_{1323} & L^{(2,3)}_{1331} & L^{(2,3)}_{1332} & L^{(2,3)}_{1333} \\ L^{(2,3)}_{2311} & L^{(2,3)}_{2312} & L^{(2,3)}_{2313} & L^{(2,3)}_{2321} & L^{(2,3)}_{2322} & L^{(2,3)}_{2323} & L^{(2,3)}_{2331} & L^{(2,3)}_{2332} & L^{(2,3)}_{2333} \\ L^{(2,3)}_{3311} & L^{(2,3)}_{3312} & L^{(2,3)}_{3313} & L^{(2,3)}_{3321} & L^{(2,3)}_{3322} & L^{(2,3)}_{3323} & L^{(2,3)}_{3331} & L^{(2,3)}_{3332} & L^{(2,3)}_{3333} \end{pmatrix}, \tag{71}$$

$$L_{ikl}^{(1,2)} = \begin{pmatrix} L_{111}^{(1,2)} & L_{112}^{(1,2)} & L_{113}^{(1,2)} & L_{121}^{(1,2)} & L_{122}^{(1,2)} & L_{123}^{(1,2)} & L_{131}^{(1,2)} & L_{132}^{(1,2)} & L_{133}^{(1,2)} \\ L_{211}^{(1,2)} & L_{212}^{(1,2)} & L_{213}^{(1,2)} & L_{221}^{(1,2)} & L_{222}^{(1,2)} & L_{223}^{(1,2)} & L_{231}^{(1,2)} & L_{232}^{(1,2)} & L_{233}^{(1,2)} \\ L_{311}^{(1,2)} & L_{312}^{(1,2)} & L_{313}^{(1,2)} & L_{321}^{(1,2)} & L_{322}^{(1,2)} & L_{323}^{(1,2)} & L_{331}^{(1,2)} & L_{332}^{(1,2)} & L_{333}^{(1,2)} \end{pmatrix}, \tag{72}$$

$$L_{ikl}^{(1,3)} = \begin{pmatrix} L_{111}^{(1,3)} & L_{112}^{(1,3)} & L_{113}^{(1,3)} & L_{121}^{(1,3)} & L_{122}^{(1,3)} & L_{123}^{(1,3)} & L_{131}^{(1,3)} & L_{132}^{(1,3)} & L_{133}^{(1,3)} \\ L_{211}^{(1,3)} & L_{212}^{(1,3)} & L_{213}^{(1,3)} & L_{221}^{(1,3)} & L_{222}^{(1,3)} & L_{223}^{(1,3)} & L_{231}^{(1,3)} & L_{232}^{(1,3)} & L_{233}^{(1,3)} \\ L_{311}^{(1,3)} & L_{312}^{(1,3)} & L_{313}^{(1,3)} & L_{321}^{(1,3)} & L_{322}^{(1,3)} & L_{323}^{(1,3)} & L_{331}^{(1,3)} & L_{332}^{(1,3)} & L_{333}^{(1,3)} \end{pmatrix}, \tag{73}$$

$$L_{ijkl}^{(3,3)} = \begin{pmatrix} L_{1111}^{(3,3)} & L_{1112}^{(3,3)} & L_{1113}^{(3,3)} & L_{1121}^{(3,3)} & L_{1122}^{(3,3)} & L_{1123}^{(3,3)} & L_{1131}^{(3,3)} & L_{1132}^{(3,3)} & L_{1133}^{(3,3)} \\ L_{1211}^{(3,3)} & L_{1212}^{(3,3)} & L_{1213}^{(3,3)} & L_{1221}^{(3,3)} & L_{1222}^{(3,3)} & L_{1223}^{(3,3)} & L_{1231}^{(3,3)} & L_{1232}^{(3,3)} & L_{1233}^{(3,3)} \\ L_{1311}^{(3,3)} & L_{1312}^{(3,3)} & L_{1313}^{(3,3)} & L_{1321}^{(3,3)} & L_{1322}^{(3,3)} & L_{1323}^{(3,3)} & L_{1331}^{(3,3)} & L_{1332}^{(3,3)} & L_{1333}^{(3,3)} \\ L_{2111}^{(3,3)} & L_{2112}^{(3,3)} & L_{2113}^{(3,3)} & L_{2121}^{(3,3)} & L_{2122}^{(3,3)} & L_{2123}^{(3,3)} & L_{2131}^{(3,3)} & L_{2132}^{(3,3)} & L_{2133}^{(3,3)} \\ L_{2211}^{(3,3)} & L_{2212}^{(3,3)} & L_{2213}^{(3,3)} & L_{2221}^{(3,3)} & L_{2222}^{(3,3)} & L_{2223}^{(3,3)} & L_{2231}^{(3,3)} & L_{2232}^{(3,3)} & L_{2233}^{(3,3)} \\ L_{2311}^{(3,3)} & L_{2312}^{(3,3)} & L_{2313}^{(3,3)} & L_{2321}^{(3,3)} & L_{2322}^{(3,3)} & L_{2323}^{(3,3)} & L_{2331}^{(3,3)} & L_{2332}^{(3,3)} & L_{2333}^{(3,3)} \\ L_{3111}^{(3,3)} & L_{3112}^{(3,3)} & L_{3113}^{(3,3)} & L_{3121}^{(3,3)} & L_{3122}^{(3,3)} & L_{3123}^{(3,3)} & L_{3131}^{(3,3)} & L_{3132}^{(3,3)} & L_{3133}^{(3,3)} \\ L_{3211}^{(3,3)} & L_{3212}^{(3,3)} & L_{3213}^{(3,3)} & L_{3221}^{(3,3)} & L_{3222}^{(3,3)} & L_{3223}^{(3,3)} & L_{3231}^{(3,3)} & L_{3232}^{(3,3)} & L_{3233}^{(3,3)} \\ L_{3311}^{(3,3)} & L_{3312}^{(3,3)} & L_{3313}^{(3,3)} & L_{3321}^{(3,3)} & L_{3322}^{(3,3)} & L_{3323}^{(3,3)} & L_{3331}^{(3,3)} & L_{3332}^{(3,3)} & L_{3333}^{(3,3)} \end{pmatrix}, \tag{74}$$

$$L^{(1,4)}_{ilmn} = \begin{pmatrix}
L^{(1,4)}_{1111} & L^{(1,4)}_{2111} & L^{(1,4)}_{3111} \\
L^{(1,4)}_{1112} & L^{(1,4)}_{2112} & L^{(1,4)}_{3112} \\
L^{(1,4)}_{1113} & L^{(1,4)}_{2113} & L^{(1,4)}_{3113} \\
L^{(1,4)}_{1121} & L^{(1,4)}_{2121} & L^{(1,4)}_{3121} \\
L^{(1,4)}_{1122} & L^{(1,4)}_{2122} & L^{(1,4)}_{3122} \\
L^{(1,4)}_{1123} & L^{(1,4)}_{2123} & L^{(1,4)}_{3123} \\
L^{(1,4)}_{1131} & L^{(1,4)}_{2131} & L^{(1,4)}_{3131} \\
L^{(1,4)}_{1132} & L^{(1,4)}_{2132} & L^{(1,4)}_{3132} \\
L^{(1,4)}_{1133} & L^{(1,4)}_{2133} & L^{(1,4)}_{3133} \\
L^{(1,4)}_{1211} & L^{(1,4)}_{2211} & L^{(1,4)}_{3211} \\
L^{(1,4)}_{1212} & L^{(1,4)}_{2212} & L^{(1,4)}_{3212} \\
L^{(1,4)}_{1213} & L^{(1,4)}_{2213} & L^{(1,4)}_{3213} \\
L^{(1,4)}_{1221} & L^{(1,4)}_{2221} & L^{(1,4)}_{3221} \\
L^{(1,4)}_{1222} & L^{(1,4)}_{2222} & L^{(1,4)}_{3222} \\
L^{(1,4)}_{1223} & L^{(1,4)}_{2223} & L^{(1,4)}_{3223} \\
L^{(1,4)}_{1231} & L^{(1,4)}_{2231} & L^{(1,4)}_{3231} \\
L^{(1,4)}_{1232} & L^{(1,4)}_{2232} & L^{(1,4)}_{3232} \\
L^{(1,4)}_{1233} & L^{(1,4)}_{2233} & L^{(1,4)}_{3233} \\
L^{(1,4)}_{1311} & L^{(1,4)}_{2311} & L^{(1,4)}_{3311} \\
L^{(1,4)}_{1312} & L^{(1,4)}_{2312} & L^{(1,4)}_{3312} \\
L^{(1,4)}_{1313} & L^{(1,4)}_{2313} & L^{(1,4)}_{3313} \\
L^{(1,4)}_{1321} & L^{(1,4)}_{2321} & L^{(1,4)}_{3321} \\
L^{(1,4)}_{1322} & L^{(1,4)}_{2322} & L^{(1,4)}_{3322} \\
L^{(1,4)}_{1323} & L^{(1,4)}_{2323} & L^{(1,4)}_{3323} \\
L^{(1,4)}_{1331} & L^{(1,4)}_{2331} & L^{(1,4)}_{3331} \\
L^{(1,4)}_{1332} & L^{(1,4)}_{2332} & L^{(1,4)}_{3332} \\
L^{(1,4)}_{1333} & L^{(1,4)}_{2333} & L^{(1,4)}_{3333}
\end{pmatrix}^{T} \tag{75}$$

By virtue of Onsager-Casimir relations (14)-(18) the sub-matrices $L^{(1,1)}_{ik}$, $L^{(2,2)}_{jikl}$, $L^{(3,3)}_{ijkl}$ and $L^{(4,4)}_{pijlmn}$ are symmetric and we can use these relations to rewrite (68) as follows

$$\{L_{\alpha\beta}\} = \left(\begin{array}{c|c|c|c} \overset{3\times3}{L^{(1,1)}_{ik}} & \overset{3\times9}{L^{(1,2)}_{ikl}} & \overset{3\times9}{L^{(1,3)}_{ikl}} & \overset{3\times27}{L^{(1,4)}_{ilmn}} \\ \hline \overset{9\times3}{L^{(1,2)}_{kji}} & \overset{9\times9}{L^{(2,2)}_{jikl}} & \overset{9\times9}{L^{(2,3)}_{jikl}} & \overset{9\times27}{L^{(2,4)}_{jilmn}} \\ \hline \overset{9\times3}{L^{(1,3)}_{kij}} & \overset{9\times9}{L^{(2,3)}_{klij}} & \overset{9\times9}{L^{(3,3)}_{ijkl}} & \overset{9\times27}{L^{(3,4)}_{ijlmn}} \\ \hline \overset{27\times3}{L^{(1,4)}_{kpij}} & \overset{27\times9}{L^{(2,4)}_{klpij}} & \overset{27\times9}{L^{(3,4)}_{klpij}} & \overset{27\times27}{L^{(4,4)}_{pijlmn}} \end{array}\right) \quad (\alpha, \beta = 1, \ldots, 48). \tag{76}$$

Furthermore, from the form (21) for the entropy production, we can write the matrix $L_{\alpha\beta}$ as follows

$$\{L_{\alpha\beta}\} = \left(\begin{array}{c|c|c|c} \overset{3\times3}{L^{(1,1)}_{ik}} & \overset{3\times9}{L^{(1,2)}_{ikl} + L^{(1,2)}_{ilk}} & \overset{3\times9}{2L^{(1,3)}_{ikl}} & \overset{3\times27}{L^{(1,4)}_{ilmn} + L^{(1,4)}_{inlm}} \\ \hline \overset{9\times3}{0} & \overset{9\times9}{L^{(2,2)}_{jikl}} & \overset{9\times9}{L^{(2,3)}_{jikl} + L^{(2,3)}_{ijkl}} & \overset{9\times27}{L^{(2,4)}_{jilmn} + L^{(2,4)}_{ijnlm}} \\ \hline \overset{9\times3}{0} & \overset{9\times9}{0} & \overset{9\times9}{L^{(3,3)}_{ijkl}} & \overset{9\times27}{L^{(3,4)}_{ijlmn} + L^{(3,4)}_{ijnlm}} \\ \hline \overset{27\times3}{0} & \overset{27\times9}{0} & \overset{27\times9}{0} & \overset{27\times27}{L^{(4,4)}_{pijlmn}} \end{array}\right) \tag{77}$$

$(\alpha, \beta = 1, \ldots, 48)$.

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