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Nonlocal effects and second sound in a nonequilibrium steady state

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The influence of weakly nonlocal effects on the speed of second sound along or against the direction of a nonvanishing average heat flow is explored in a formalism based on a dynamical nonequilibrium temperature. The restrictions from the second law of thermodynamics on the nonlocal evolution equation of this temperature are studied from a gradient extension of classical Liu procedure, and the corresponding form of the entropy is derived. Stability requirements obtained from the second differential of the entropy are seen to restrict a nonlinear dependence of the thermal conductivity on the temperature gradient.

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I. INTRODUCTION

Interest in generalized transport equations incorporating memory, nonlocal, and nonlinear effects has fostered the exploitation of the form of the second law of thermodynamics.[1–3] For instance, compatibility of generalized equations with the second law often requires the use of generalized entropies going beyond the local-equilibrium entropy.[1,4,5] On the other side, some well-known methods for the exploitation of the second law, as the classical Liu method,[6,7] must also be generalized to incorporate different restrictions consistent with the higher order of transport equations.[8] Here, we will deal with weakly nonlocal effects in heat transport, a topic of much interest in small systems, where small temperature differences divided by a small-scale length may lead to very high temperature gradients.[9] Furthermore, it is known that there is a finite build-up time for the appearance of a thermal current, after that a temperature gradient is suddenly clamped, of the order of magnitude of some fractions of microseconds.[10,11] Though this value may seem small, it may be relevant for microdevices working at high frequencies in computers.

Of course, second sound and other related phenomena cannot be described by the classical Fourier equation since it leads to infinite speed of propagation for thermal disturbances,[12] and therefore more general heat transport equations must be looked for.[4,5,10,11] In a series of recent papers,[13–15] a thermodynamic description of relaxational heat transfer based on the so-called semiempirical temperature scale has been employed. The essential idea of this approach is to assume that the heat flux is given by

$$\mathbf{q} = -\kappa \nabla \beta, \tag{1}$$

with κ as a suitable function of thermodynamic state variables representing the thermal conductivity and β a semiempirical temperature.

According to Fourier law, Eq. (1) preserves the assumption that the heat flux should be in the inverted direction of the gradient of a potential function, but in contrast to it, β is a dynamical nonequilibrium temperature which differs from thermodynamic absolute temperature θ. The semiempirical temperature β is related to the absolute one by a kinetic equation of the form

$$\dot{\beta} = -\frac{1}{\tau}(\beta - \theta), \tag{2}$$

where τ is a relaxation time characterizing the approach of β toward θ. In some occasions, β has been also called a dynamical temperature because its difference with the thermodynamic temperature θ has a dynamical origin according to Eq. (2). By design, at equilibrium, β is a suitable regular function of local-equilibrium temperature T. Conditions under which θ reduces to T are shown in Refs. 16 and 17. In Sec. II the differences among β, θ, ant T will be further discussed and clarified.

When Eq. (1) is combined with Eq. (2), if the relaxation time τ and the thermal conductivity κ are supposed to be constant, one obtains

$$\tau \dot{\mathbf{q}} + \mathbf{q} = -\kappa \nabla \theta, \tag{3}$$

which is the well-known Maxwell-Cattaneo equation,[1,4,5,12] leading to a finite speed for thermal signals and to the existence of a propagative temperature wave known as second sound.

The aim of the present paper is twofold: (1) to revisit the description of Eq. (3) in terms of a dynamical semiempirical temperature and its gradient on purely thermodynamic grounds in order to gain further insight on the thermodynamic aspects of nonlocal terms in transport equation and (2) to apply the theory to describe the effects of a nonvanishing steady heat flux on the propagation of second sound. This may be of practical interest for the analysis of the speed of signals in nonequilibrium steady states. These topics are of much interest in the intermediate regime between diffusive and ballistic heat transports, which is one of the hot topics in current transport theory because of its implications in nanotechnology.

In more detail, in Sec. II we show how it is possible to define a semiempirical temperature β by revisiting the classical Cattaneo approach[12] to the problem of finite speed of propagation of thermal disturbances.[13] We also point out the experimental conditions under which the heat conduction law (1) is true. In Sec. III we consider a rigid body for which the constitutive variables are the specific internal energy e and the semiempirical temperature β, together with their gradients ∇e and $\nabla\beta$, introduced in order to describe nonlocal effects which are lacking in Eq. (2).

Section IV explores the generalized heat transport model in the framework of weakly nonlocal extended thermodynamics. After having exploited the second law of thermodynamics by applying an extended Liu procedure,[8] we discuss the consequent thermodynamic restrictions leading to a nonlocal entropy function and point out the conditions under which hyperbolic heat transport can be found.

In Sec. V, for a one-dimensional case, we investigate the compatibility of the introduced form of entropy function with the principle of maximum entropy at equilibrium states. Furthermore, conditions for the convexity of entropy functions[4,5,18] outside the equilibrium state are discussed, and their connections with the stability of nonequilibrium steady states are pointed out. This section is specially interesting for nonlinear heat transfer, as some of the stability conditions may be related to nonlinear contributions to a generalized thermal conductivity.

Section VI is devoted to compare the results of this paper both with the previous results obtained in the extended irreversible thermodynamics (EIT) (Refs. 1 and 4) and with the Onsager approach,[19,20] while in Sec. VII concluding remarks together with possible further developments are presented.

II. DYNAMICAL, NONEQUILIBRIUM, AND LOCAL-EQUILIBRIUM TEMPERATURE

Classical irreversible thermodynamics lays on the so-called local-equilibrium hypothesis,[21] according to which it is assumed that the concept of entropy, which is well defined for processes close to the equilibrium, does not need a reformulation out of equilibrium, as it can be naturally extended, locally, to nonhomogeneous nonequilibrium situations. If the hypothesis of local equilibrium is relaxed, then one is faced with the problem of defining entropy in nonequilibrium conditions. One of the possible ways for circumventing the problem is to consider the entropy as a primitive quantity.[22] In any case, once a constitutive equation for the entropy is given, the thermodynamic absolute temperature θ can be obtained by the thermodynamic restriction,[23]

$$\theta^{-1} = \frac{\partial s}{\partial e}, \tag{4}$$

with s as the specific entropy and e as the specific internal energy. On the other hand, beside equilibrium quantities, s contains nonequilibrium variables too, such as internal variables in rational thermodynamics[24] or dissipative fluxes in extended thermodynamics.[1,4,5] Hence, we are led to the natural conclusion that the function θ, as defined by Eq. (4), is a truly nonequilibrium quantity. Furthermore, the local-equilibrium temperature T can be obtained from the right-hand side of Eq. (4) by letting the nonequilibrium variables vanish. For instance, in extended irreversible thermodynamics, the entropy of rigid heat conductors takes the form[25]

$$s(e;\mathbf{q}) = s_{\text{eq}}(e) - \frac{\tau}{2\kappa T^2}\mathbf{q}\cdot\mathbf{q}, \tag{5}$$

where $s_{\text{eq}}(e)$ is the local-equilibrium entropy, while κ and τ are the heat conductivity and the relaxation time, respectively. Similar expressions can be derived in rational thermodynamics.[24,26,27]

A further problem arising in nonequilibrium thermodynamics and involving the physical properties of θ is the infinite speed of propagation of thermal disturbances whenever the classical proportionality law between heat flux and gradient of temperature is assumed to hold.[12] In fact, let us consider a rigid heat conductor, and let us suppose that its thermodynamic state space is spanned by the thermodynamic absolute temperature θ together with its gradient. In the absence of heat sources, the local balance of energy reads

$$\dot{e} + q_{i,k}\delta_{ik} = 0, \tag{6}$$

where q_i, with $i=(1,2,3)$, is the ith component of the heat flux vector[2,3] and $\delta_{ik}=\delta_{ki}$ is the Kronecker symbol. Now and in what follows, Einstein's convention of summation over repeated indices will be used. Moreover, the subscript $(,k)$ denotes the partial derivatives with respect to the coordinate x_k, and a superposed dot stands for the Lagrangian time derivative.

By postulating the linear constitutive equations

$$e = c_v\theta, \quad q_i = -\kappa\theta_{,i} \tag{7}$$

once the specific heat c_v and the thermal conductivity κ are supposed to be constant, from Eq. (6) we obtain the classical diffusion equation,

$$\dot{\theta} = \frac{\kappa}{c_v}\theta_{,ii}. \tag{8}$$

Let us consider now the following initial value problem:

$$\dot{\theta} = \frac{\kappa}{c_v}\frac{\partial^2\theta}{\partial x^2},$$

$$\theta(x\neq 0;0) = 0, \quad \theta(0;0) = 1 \tag{9}$$

for a semi-infinite wire represented by the non-negative real axis $x \geq 0$. The fundamental solution of Eq. (9) is[28]

$$\theta(x;t) = \frac{1}{2\sqrt{\dfrac{\pi\kappa t}{c_v}}}e^{-x^2/4(\kappa/c_v)t}. \tag{10}$$

From Eq. (10) it follows that $\theta(\bar{x};t)>0$ for arbitrarily small t and for $\bar{x}$ as large as we wish. Such a property is known in the literature as *paradox of infinite speed of propagation of thermal disturbances*.[5,12] In order to remove such a paradox, in 1948 Cattaneo[12] proposed his celebrated evolution [Eq. (3)] for the heat flux. From Eqs. (3) and (6) it follows

$$\tau\ddot{\theta} + \dot{\theta} = \frac{\kappa}{c_v}\theta_{,ii}, \tag{11}$$

which is the well-known telegraphic equation for the evolution of the temperature.[12] For the semi-infinite wire considered above, it reduces to

$$\tau\ddot{\theta} + \dot{\theta} = \frac{\kappa}{c_v}\frac{\partial^2\theta}{\partial x^2}, \tag{12}$$

allowing the propagation of temperature pulses with finite speed

$$U_0 = \sqrt{\frac{\kappa}{\tau c_v}}.$$

Nevertheless, Cattaneo's proposal (3) cannot be considered as an exhaustive solution of the problem because of the following considerations: (i) the compatibility of Eq. (3) with the basic principles of continuum thermodynamics must be proved and (ii) if the material functions τ, κ, and c_v depend on the temperature,[29] the applied procedure could no longer give rise to a hyperbolic equation. In literature one can find several different models which have been designed to be in accordance with nonequilibrium thermodynamics and still leading to Eq. (3).[10,30,31]

As an example, let us mention the approach of extended irreversible thermodynamics.[1,4] In such a theory the state space of rigid heat conductors is spanned by the internal energy e, ruled by Eq. (6), by the heat flux $\mathbf{q}$, and by the flux of heat flux $\mathbf{Q}$. Both the dissipative fluxes $\mathbf{q}$ and $\mathbf{Q}$ are governed by the following balance laws:

$$\tau\dot{q}_i + Q_{ij_{,j}} = \sigma_i, \tag{13}$$

$$\tau_1\dot{Q}_{ij} + P_{ijk_{,k}} = \Sigma_{ij}, \tag{14}$$

where $\mathbf{P}$ is the flux of $\mathbf{Q}$, σ and $\boldsymbol{\Sigma}$ are the productions of $\mathbf{q}$ and $\mathbf{Q}$, respectively, while τ_1 is a relaxation time. The previous system is closed by assigning suitable constitutive equations for the flux $\mathbf{P}$ and for the productions σ and $\boldsymbol{\Sigma}$ as functions of the set of thermodynamic variables $\{e;\mathbf{q};\mathbf{Q}\}$. For regular solutions, one can consider the case in which τ_1 is negligible, and so the divergence of $\mathbf{Q}$ can be calculated by Eq. (14) and substituted into Eq. (13). That way, a general governing equation for $\mathbf{q}$, reducing to Eq. (3) in particular cases, can be obtained.

A different approach, starting from Cattaneo's original derivation[12] of Eq. (3), lays on the definition of a new dynamical temperature.[13] Let us give below a short sketch of this approach, namely, the definition of β and its main properties (see Ref. 13 for more detail). To this end let us consider a gas in macroscopic mechanical equilibrium and let G denote the average of the kinetic energy of the molecules. Moreover, let us consider a steady state, in which G is independent of time and constant on the family of planes orthogonal to a given direction r. Then, G can be supposed to depend on the position x of the points of r only. Under such a hypothesis, it can be easily proved[12] that the net flux of G across the generic plane at x is

$$q = \int_0^\infty \frac{-\lambda_c c}{3}\frac{\partial G}{\partial x}dN_c, \tag{15}$$

where c denotes the speed of the molecules, λ_c is their mean-free path, while dN_c is the number of molecules whose velocity belongs to the interval $[c;c+dc]$. On the other hand, from the relation $G=\frac{3}{2}k_B\theta$, with k_B as the Boltzmann constant, we easily get from Eq. (15) the classical Fourier law,

$$q = -\kappa\frac{\partial\theta}{\partial x}, \tag{16}$$

with the heat conductivity κ given by

$$\kappa = \int_0^\infty \frac{\lambda_c c k_B}{2}dN_c. \tag{17}$$

If we go beyond the stationary state, then G has to be considered to depend on space and time. A different flux of G can now be carried out by expanding G up to the second order. In such a case

$$q = \int_0^\infty \left(\frac{-\lambda_c c}{3}\frac{\partial G}{\partial x} + \frac{l_c}{3}\frac{\partial^2 G}{\partial x\,\partial t}\right)dN_c, \tag{18}$$

with l_c denoting the mean square of the mean-free path of the molecules.[12] Then, one can write

$$q = -\kappa\frac{\partial\theta}{\partial x} + \sigma\frac{\partial^2\theta}{\partial x\,\partial t}, \tag{19}$$

with

$$\sigma = \int_0^\infty \frac{l_c k_B}{2}dN_c. \tag{20}$$

Now, let us define the dynamical temperature[13]

$$\beta = \theta - \tau\dot{\theta}, \tag{21}$$

where $\tau=\frac{\sigma}{\kappa}$ is a small relaxation time whose order of magnitude in some cases, such as second-sound propagation at low temperature, is in the interval $[10^{-12}\ \text{s};\ 10^{-9}\ \text{s}]$.[29] From the physical point of view β is different from θ, since it contains a frictional term related to the time rate of θ. Such a term introduces hyperbolicity since it reduces the speed of propagation of β through the gas.

In order to obtain the evolution equation for β, let us derive Eq. (21) with respect to time. This yields

$$\dot{\beta} = \dot{\theta} - \tau\ddot{\theta}. \tag{22}$$

Then, we calculate $\dot{\theta}$ from Eq. (21) and substitute it into Eq. (22). That way, we are led to

$$\tau\dot{\beta} = \theta - \beta - \tau^2\ddot{\theta}. \tag{23}$$

Finally, if we consider only regular solutions of Eq. (23), then the smallness of τ ensures that we can neglect the last term at the right-hand side of Eq. (23), getting so

$$\tau\dot{\beta} + \beta = \theta. \tag{24}$$

From the mathematical point of view we see that β is related to θ by a linear differential equation, needing suitable initial conditions to be solved. More generally, we can allow β to reduce to a function of θ at the equilibrium, which is tantamount to measure β in a different scale. To this end we can generalize Eq. (24) as follows:

$$\dot{\beta} = f(\theta;\beta), \tag{25}$$

where the form of f can be determined by second-sound measurements.[14]

It is worth observing that the equilibrium of β (i.e., $\dot{\beta}=0$) should not be confused with the thermodynamic equilibrium of the system. When β is in equilibrium, it reduces to a function of the nonequilibrium absolute temperature θ and can be determined analytically as the solution of

$$f(\theta;\beta) = 0. \tag{26}$$

According to the terminology of classical irreversible thermodynamics, β can be regarded as an internal state variable.[32] In a sense, its definition is analytical since it is given by Eq. (25). However, such an equation can be solved only if the form of f and the initial condition are known, and such a knowledge requires suitable experiments. These considerations motivated the name of semiempirical temperature for β.[13] From the operational point of view, β can be considered to be in equilibrium at a characteristic time scale with respect to which τ is negligible.

From relation (19) we get the Fourier-type heat conduction law,

$$q = -\kappa\frac{\partial\beta}{\partial x}. \tag{27}$$

Finally, due to Eqs. (24) and (27), the local balance of energy (6) yields for β the telegraphic equation,

$$\tau\ddot{\beta} + \dot{\beta} = \frac{k}{c_v}\frac{\partial^2\beta}{\partial x^2}, \tag{28}$$

leading to finite speeds of propagation of thermal disturbances. A different situation arises if we generalize Eq. (27) by assuming that q depends not only on both temperatures but also on both gradients, namely,

$$q = -\kappa_1\frac{\partial\theta}{\partial x} - \kappa\frac{\partial\beta}{\partial x}. \tag{29}$$

We will show in Sec. VI that the expression above follows by the Onsager approach to nonequilibrium thermodynamics.[21] Due to the presence of the gradient of θ, Eq. (29) leads to a parabolic system.[33,34] However it is capable to reproduce the hyperbolic behavior if κ_1 is negligible. In order to illustrate in more detail the experimental conditions under which this can happen, let us summarize the most important properties of heat conduction in solids near absolute zero. From the microscopic point of view this phenomenon can be modeled through the phonon gas hydrodynamics.[35,36] In a solid crystal at low temperature the phonons form a rarefied gas, whose kinetic equation can be derived similarly to that of an ordinary gas. Moving through the crystal lattice they undergo two different types of collisions: (i) normal (N) collisions, which conserve the phonon momentum, and (ii) resistive (R) collisions, in which the phonon momentum is not conserved.

The frequencies ν_N and ν_R of normal and resistive collisions, respectively, determine the characteristic relaxation times $\tau_N=\frac{1}{\nu_N}$ and $\tau_R=\frac{1}{\nu_R}$. Non-Fourier diffusive heat transport takes over when ν_R tends to infinity, and hence $\tau_R=\frac{1}{\nu_R}$ tends to zero. If instead $\tau_N=\frac{1}{\nu_N}$ tends to zero, propagation of heat waves may occur. If both relaxation times are not negligible, one obtains the so-called diffusive-hyperbolic behavior, in which the regularization of the profile of the thermal pulses and the transition to the diffusive regime can be observed. Finally, the Fourier regime is restored if both relaxation times vanish.

According to the symbology of the present paper, τ_R should be identified with τ. Moreover, the considerations above suggest that κ_1 should be expressed as a function of τ_N. By assuming a linear dependency, namely, $\kappa_1=\chi\tau_N$, with χ as a suitable constant, then we get

$$q = -\chi\tau_N\frac{\partial\theta}{\partial x} - \kappa\frac{\partial\beta}{\partial x}. \tag{30}$$

The hyperbolic behavior described by the constitutive Eq. (27) takes place when $\tau\neq 0$ and $\tau_N=0$; if instead both relaxation times are not negligible, one obtains the diffusive-hyperbolic behavior.[33,34] Finally, the Fourier diffusive regime is recovered as both relaxation times approach zero.

Let us close this section by summarizing the results above. We have defined the following three different temperatures: (1) the local-equilibrium absolute temperature T, which can be defined through the second law of thermodynamics in homogeneous equilibrium situations as $\frac{1}{T}=\frac{\partial s_{\mathrm{eq}}}{\partial e}$; (2) the nonequilibrium absolute temperature θ, defined by Eq. (4), once a suitable constitutive equation for the nonequilibrium entropy s has been achieved. Since s is different from s_{eq} [see, for instance, Eq. (5)], θ will differ from T except at equilibrium; and (3) the semiempirical dynamical temperature β governed by the ordinary differential Eq. (25), which is different from θ at time scales comparable with the order of magnitude of τ but reduces to a function of θ at time scales at which τ is negligible.

Here we will be interested in fast phenomena, of time scales smaller than τ (for which β is different from θ), and in the presence of high fluxes (for which θ is different from T). In equilibrium, β, θ, and T become identical but in fast and far-from equilibrium systems they differ from each other. In what follows we look for a generalization of Eq. (2) under the assumption of non-negligible relaxation time.

III. BALANCE EQUATIONS AND ENTROPY INEQUALITY

Let us consider a rigid body whose thermodynamic state space is spanned by the specific internal energy e, the semiempirical temperature β, and their first gradients ∇e and $\nabla\beta$. The considerations developed in Sec. II suggest that in an equilibrium system, where the internal energy e is a function of the thermodynamic local-equilibrium absolute temperature T, the use of both e and β would be redundant. However, out of equilibrium, where the internal energy e is not distributed as in equilibrium, β is a truly independent quantity and not redundant with e. Furthermore, it has its own evolution equation, namely, Eq. (2) or more general versions.

The presence of the gradients of basic variables in the thermodynamic state space is introduced in view of a weakly nonlocal description. The thermodynamic analysis of such a model, when instead of β the heat flux $\mathbf{q}$ is used as an additional independent variable, was developed by Lebon *et al.*[9] by applying the standard Liu procedure.[7]

According to the chain rule, the local balance of the specific internal energy reads

$$\dot{e}+\frac{\partial q_i}{\partial e}e_{,i}+\frac{\partial q_i}{\partial \beta}\beta_{,i}+\frac{\partial q_i}{\partial e_{,k}}e_{,ki}+\frac{\partial q_i}{\partial \beta_{,k}}\beta_{,ki}=r, \tag{31}$$

with r as the specific-heat supply. As a generalization of Eq. (2) we suppose

$$\dot{\beta}=f(e;\beta;e_{,k};\beta_{,k}), \tag{32}$$

where f is a regular function of the indicated arguments, whose form must be determined through thermodynamic restrictions. Nonlocal effects appear through the gradients of e and β in the evolution Eq. (32).

The equations characterizing the process must be restricted by the second law of thermodynamics, which takes the local form

$$\frac{\partial s}{\partial e}\dot{e}+\frac{\partial s}{\partial \beta}\dot{\beta}+\frac{\partial s}{\partial e_{,i}}\dot{e}_{,i}+\frac{\partial s}{\partial \beta_{,i}}\dot{\beta}_{,i}+\frac{\partial \Phi_i^{(s)}}{\partial e}e_{,i}+\frac{\partial \Phi_i^{(s)}}{\partial \beta}\beta_{,i} +\frac{\partial \Phi_i^{(s)}}{\partial e_{,k}}e_{,ki}+\frac{\partial \Phi_i^{(s)}}{\partial \beta_{,k}}\beta_{,ki}\geq \Pi^{(s)}, \tag{33}$$

where s denotes the specific entropy $s(e;\beta;e_{,k};\beta_{,k})$, $\Pi^{(s)}$ is the entropy supply, and $\Phi_i^{(s)}$ is the ith component of the entropy flux.

One of the basic postulates of rational thermodynamics[23] is that the entropy flux is proportional to the heat flux, namely,

$$\Phi_i^{(s)}=\frac{1}{\theta}q_i, \tag{34}$$

whereas the nonequilibrium thermodynamic temperature is described, as we already observed in Sec. II, by the function

$$\theta^{-1}(e;\beta;e_{,k};\beta_{,k})=\frac{\partial s}{\partial e}. \tag{35}$$

Assumption (34) has been found to be appropriate for a very large class of phenomena, although it fails in describing the entropy flux in mixtures, and in general, in all those situations in which a diffusion of matter takes place. As a possible remedy, Müller[37,38] proposed his celebrated entropy extra flux $\mathbf{k}$, accounting for the additional flux due to matter diffusion. Recently, some generalizations of Müller's proposal have been derived by other authors, either in the framework of classical irreversible thermodynamics[39,40] or in the realm of extended irreversible thermodynamics.[9,41] All these generalizations contain Eq. (34) as a particular case. On the other hand, for the systems considered in the present paper (dielectric crystals at low temperature or nanodevices) the thermal effects are predominant so that Eq. (34) can be considered as a good approximation of the entropy flux.

As far as the form of the heat flux is concerned, since we are interested to study only the hyperbolic behavior, we suppose that Eq. (1) represents a suitable approximation of the heat flux, namely,

$$q_i=-\kappa(e;\beta)\beta_{,i}. \tag{36}$$

Due to constitutive relation (34), inequality (33) can be also rewritten as

$$\theta\left(\frac{\partial s}{\partial e}\dot{e}+\frac{\partial s}{\partial \beta}\dot{\beta}+\frac{\partial s}{\partial e_{,i}}\dot{e}_{,i}+\frac{\partial s}{\partial \beta_{,i}}\dot{\beta}_{,i}\right)+\frac{\partial q_i}{\partial e}e_{,i}+\frac{\partial q_i}{\partial \beta}\beta_{,i}+\frac{\partial q_i}{\partial e_{,k}}e_{,ki} +\frac{\partial q_i}{\partial \beta_{,k}}\beta_{,ki}-\frac{1}{\theta}q_i\left(\frac{\partial \theta_i}{\partial e}e_{,i}+\frac{\partial \theta_i}{\partial \beta}\beta_{,i}+\frac{\partial \theta_i}{\partial e_{,k}}e_{,ki}+\frac{\partial \theta_i}{\partial \beta_{,k}}\beta_{,ki}\right) \geq \theta\Pi^{(s)}. \tag{37}$$

IV. SEMIEMPIRICAL TEMPERATURE AND NONLOCAL HEAT TRANSPORT

In the present section we explore which kind of nonlocal effects appear in the introduced model (32) for the evolution of β. Moreover, we deal with generalized heat transport models inside the framework of weakly nonlocal extended thermodynamics[1] to illustrate and understand how the dynamics of the semiempirical temperature, reflected in Eq. (32), is connected with nonlocal effects in heat transport.

In order to investigate the compatibility of this equation with the second law of thermodynamics (33), we apply an extended Liu procedure.[8] In fact, in weakly nonlocal extended thermodynamics the entropy function may present some physical properties (i.e., entropy must be nonlocal) which cannot be evidenced by the direct application of the classical Liu procedure.[7] Thus, it is important to explore more general procedures enabling to grasp subtle aspects originating from nonlocality.

The extended Liu procedure consists of forming a general inequality by adding to the entropy inequality not only a linear combination of the governing equations of the wanted fields but also of their extensions up to the order of the spatial derivatives entering the constitutive equations. The factors multiplying the field equations and their extensions are Lagrange multipliers, whose physical role will be explored. Thermodynamic restrictions on the constitutive functions are obtained by an analytical procedure which starts by the observation that this inequality is linear in the highest derivatives of the wanted fields, which are considered to be completely arbitrary. Thus, we will also take into account the gradient extension of Eq. (31), reading

$$\dot{e}_{,i}+\left(\frac{\partial^2 q_k}{\partial e^2}e_{,i}+\frac{\partial^2 q_k}{\partial \beta\, \partial e}\beta_{,i}+\frac{\partial^2 q_k}{\partial e_{,j}\, \partial e}e_{,ji}+\frac{\partial^2 q_k}{\partial \beta_{,j}\, \partial e}\beta_{,ji}\right)e_{,k} +\frac{\partial q_k}{\partial e}e_{,ki}+\left(\frac{\partial^2 q_k}{\partial e\, \partial \beta}e_{,i}+\frac{\partial^2 q_k}{\partial \beta^2}\beta_{,i}+\frac{\partial^2 q_k}{\partial e_{,j}\, \partial \beta}e_{,ji} +\frac{\partial^2 q_k}{\partial \beta_{,j}\, \partial \beta}\beta_{,ji}\right)\beta_{,k}+\frac{\partial q_k}{\partial \beta}\beta_{,ki}+\left(\frac{\partial^2 q_k}{\partial e\, \partial e_{,j}}e_{,i}+\frac{\partial^2 q_k}{\partial \beta\, \partial e_{,j}}\beta_{,i}\right.$$

$$+\frac{\partial^2 q_k}{\partial e_{,z}\,\partial e_{,j}}e_{,zi}+\frac{\partial^2 q_k}{\partial \beta_{,z}\,\partial e_{,j}}\beta_{,zi}\Bigg)e_{,ki}+\frac{\partial q_k}{\partial e_{,j}}e_{,kji}$$
$$+\left(\frac{\partial^2 q_k}{\partial e\,\partial \beta_{,j}}e_{,i}+\frac{\partial^2 q_k}{\partial \beta\,\partial \beta_{,j}}\beta_{,i}+\frac{\partial^2 q_k}{\partial e_{,z}\,\partial \beta_{,j}}e_{,zi}\right.$$
$$\left.+\frac{\partial^2 q_k}{\partial \beta_{,z}\,\partial \beta_{,j}}\beta_{,zi}\right)\beta_{,ki}+\frac{\partial q_k}{\partial e_{,j}}\beta_{,kji}+\frac{\partial r}{\partial e}e_{,i}+\frac{\partial r}{\partial \beta}\beta_{,i}$$
$$+\frac{\partial r}{\partial e_{,k}}e_{,ki}+\frac{\partial r}{\partial \beta_{,k}}\beta_{,ki}=0, \tag{38}$$

together with the gradient extension of the evolution Eq. (32) for β, namely,

$$\dot{\beta}_{,i}=\frac{\partial f}{\partial e}e_{,i}+\frac{\partial f}{\partial \beta}\beta_{,i}+\frac{\partial f}{\partial e_{,k}}e_{,ki}+\frac{\partial f}{\partial \beta_{,k}}\beta_{,ki}. \tag{39}$$

Let us note that in writing the balances above we have considered the general case in which the heat flux depends on the whole set of state variables. That way the extension of the classical Liu procedure will result clearer.

A. Hyperbolic heat transport

In order to take into account the second law of thermodynamics, we substitute into Eq. (33) constraints (31) and (32), together with their gradient extensions (38) and (39). It yields

$$\dot{s}+\Phi^{(s)}_{i_{,k}}\delta_{ik}-\Pi^{(s)}-\lambda^{(e)}(\dot{e}+q_{i_{,k}}\delta_{ik}-r)-\lambda^{(\beta)}(\dot{\beta}-f)$$
$$-\lambda_i^{(e)}(\dot{e}_{,i}+q_{k_{,ji}}\delta_{jk}-r_{,i})-\lambda_i^{(\beta)}(\dot{\beta}_{,i}-f_{,i})\geq 0, \tag{40}$$

where, for the sake of simplicity, we adopted a compact notation, denoting with $q_{k_{,ki}}$ the ith component of $\nabla(\nabla\cdot\mathbf{q})$.

In the extended Liu inequality (40) the terms $\lambda^{(e)}$, $\lambda^{(\beta)}$, $\lambda_i^{(e)}$, and $\lambda_i^{(\beta)}$ are suitable Lagrange multipliers which allow us to incorporate the corresponding restrictions coming from Eqs. (31), (32), (38), and (39). The introduction of the gradient extensions by means of $\lambda_i^{(e)}$ and $\lambda_i^{(\beta)}$ represents a different approach to the exploitation of second law[8] since the standard point of view would have been to impose only the evolution equations of state space variables through $\lambda^{(e)}$ and $\lambda^{(\beta)}$.[7] The gradient extensions are specially relevant in the context of nonlocal effects, as it will be seen below.

According to Ref. 8 and due to Eq. (36), by inequality (40) the following set of thermodynamic restrictions can be derived:

$$\frac{\partial s}{\partial e}=\lambda^{(e)}, \tag{41}$$

$$\frac{\partial s}{\partial e_{,i}}=\lambda_i^{(e)}, \tag{42}$$

$$\frac{\partial s}{\partial \beta_{,i}}=\lambda_i^{(\beta)}, \tag{43}$$

$$\frac{1}{\theta^2}\frac{\partial \theta}{\partial e_{,k}}q_i-\lambda_i^{(\beta)}\frac{\partial f}{\partial e_{,k}}+\lambda_i^{(e)}\left(\frac{\partial q_k}{\partial e}-\frac{\partial r}{\partial e_{,k}}\right)=0, \tag{44}$$

$$\frac{1}{\theta^2}\frac{\partial \theta}{\partial \beta_{,k}}q_i-\lambda_i^{(\beta)}\frac{\partial f}{\partial \beta_{,k}}+\lambda_i^{(e)}\left(\frac{\partial q_k}{\partial \beta}-\frac{\partial r}{\partial \beta_{,k}}\right)+\lambda_i^{(e)}\left(\frac{\partial^2 q_j}{\partial \beta_{,k}\,\partial e}e_{,j}\right.$$
$$\left.+\frac{\partial^2 q_j}{\partial \beta_{,k}\,\partial \beta}\beta_{,j}\right)+\lambda_j^{(e)}\left(\frac{\partial^2 q_i}{\partial e\,\partial \beta_{,k}}e_{,j}+\frac{\partial^2 q_j}{\partial \beta\,\partial \beta_{,k}}\beta_{,j}\right)=0, \tag{45}$$

$$\lambda_i^{(e)}\frac{\partial q_j}{\partial \beta_{,k}}=0. \tag{46}$$

Now, let us focus for a while our attention on relation (46). It is satisfied only in the trivial case, namely, either

$$\lambda_i^{(e)}=0 \tag{47}$$

or

$$\frac{\partial q_j}{\partial \beta_{,k}}=0. \tag{48}$$

Due to the constitutive assumption (36), it follows that Eq. (47) must hold. Furthermore, taking into account Eq. (46), the thermodynamic restrictions (44) and (45) yield

$$\frac{\partial f}{\partial e_{,k}}=\frac{1}{3\lambda_i^{(\beta)}}\frac{1}{\theta^2}q_i\frac{\partial \theta}{\partial e_{,k}}, \tag{49}$$

$$\frac{\partial f}{\partial \beta_{,k}}=\frac{1}{3\lambda_i^{(\beta)}}\frac{1}{\theta^2}q_i\frac{\partial \theta}{\partial \beta_{,k}}, \tag{50}$$

$$\frac{\partial s}{\partial \beta}f+\left(\lambda_i^{(\beta)}\frac{\partial f}{\partial e}-\frac{1}{\theta^2}q_i\frac{\partial \theta}{\partial e}\right)e_{,i}+\left(\lambda_i^{(\beta)}\frac{\partial f}{\partial \beta}-\frac{1}{\theta^2}q_i\frac{\partial \theta}{\partial \beta}\right)\beta_{,i}\geq 0, \tag{51}$$

if the classical assumption of rational thermodynamics $\Pi^{(s)}=\frac{\partial s}{\partial e}r$ holds.

From relations (41)–(43) and (46), it turns out that the specific entropy s is nonlocal with respect to semiempirical temperature β but not with respect to the specific internal energy e. Thus, a suitable form of specific entropy up to the second order in $\beta_{,i}$ may be

$$s(e;\beta;\beta_{,k})=s_0(e;\beta)-\frac{1}{2}s_{\beta}(e;\beta)\beta_{,i}\beta_{,i}, \tag{52}$$

where $s_0(e;\beta)$ is the part of the entropy which does not depend on the gradient of β, while $s_{\beta}(e;\beta)$ is a regular scalar function. This term vanishes at equilibrium, where $\beta_{,i}=0$. Equation (52) is in accordance with the representation theorems of isotropic scalar functions, and moreover, it ensures that the principle of maximum entropy at the equilibrium is satisfied,[42] as we will see below when, in pointing out the stability conditions, restrictions on the admissible values of $s_{\beta}(e;\beta)$ will arise.

Along with the thermodynamic restriction (43), form (52) yields

$$\lambda_i^{(\beta)}=-s_{\beta}(e;\beta)\beta_{,i}. \tag{53}$$

Note that without the gradient extension proposed in Ref. 8, the entropy would have been predicted to be independent of $\beta_{,i}$. In Sec. IV B we will explore the physical consequences of $s_\beta(e;\beta)$.

If one identifies the nonequilibrium thermodynamic temperature with Eq. (35), once relation (52) holds, it is easy to see that $\theta=\theta(e;\beta;\beta_{,i})$, and then from Eq. (49) it follows that the evolution equation for the dynamical semiempirical temperature β is not dependent on the gradient of the internal energy e, i.e., $f=f(e;\beta;\beta_{,i})$. Thus, from Eq. (50), with straightforward calculations and integrating with respect to $\beta_{,i}$, we finally get

$$f(e;\beta;\beta_{,k})=f_0(e;\beta)+\frac{1}{2}f_1(e;\beta)\beta_{,i}\beta_{,i}, \tag{54}$$

where f_0 is a suitable function of the indicated arguments and

$$f_1(e;\beta)=\frac{\kappa}{s_\beta}\frac{\partial s_\beta}{\partial e}. \tag{55}$$

Looking at relation (2), between the semiempirical temperature and the absolute one, and definition (32) of the function f, we may suppose

$$f_0(e;\beta)=-\frac{1}{\tau}(\beta-\theta). \tag{56}$$

In writing Eq. (56) we have also taken into account that the specific internal energy e and the absolute temperature θ are related by means of the specific heat c_v, as $c_v=\frac{\partial e}{\partial \theta}$.

B. Second sound in the presence of a heat flux

Here, we will study the consequences of the new weakly nonlocal term appearing in Eq. (54), which is the main difference between Eqs. (2) and (54). Taking into account relations (54)–(56), for constant material functions, gradient extension (39) of the evolution equation for β leads to

$$\dot{\beta}_{,i}=-\frac{1}{\tau}(\beta_{,i}-\theta_{,i})+f_1\beta_{,k}\beta_{,ki}, \tag{57}$$

where, as in what follows in this section, we have neglected the third-order terms with respect to $e_{,i}$ and $\beta_{,i}$. Combining Eq. (57) with the constitutive assumption (36), we obtain

$$\tau\dot{q}_i+q_i=-\kappa\theta_{,i}-\frac{\tau f_1}{\kappa}q_k q_{k_{,i}}. \tag{58}$$

The evolution Eq. (58) for the heat flux q_i reduces to the Maxwell-Cattaneo Eq. (3) when $f_1(e;\beta)=0$. We want to stress that in the classical Liu procedure for the exploitation of the second law,[7] the entropy would have been predicted to be independent of $\beta_{,i}$, namely, $s_\beta(e;\beta)=0$, and thus $f_1(e;\beta)=0$.

For the sake of simplicity, now let us restrict ourselves to the problem of heat transport in a one-dimensional rigid body. For a high-frequency perturbation δ around a steady state under a heat flux, from Eq. (58) it follows that

$$\delta\dot{q}_x+\frac{1}{\tau}\delta q_x=-\frac{\kappa}{\tau}(\delta\theta)_{,x}-\frac{f_1}{\kappa}q_{x_0}(\delta q_x)_{,x}, \tag{59}$$

where $q_{x_0}=-\kappa\theta_{0_{,x}}$ and θ_0 are, respectively, the heat flux and the thermodynamic absolute temperature at the steady state, and x denotes the longitudinal spatial coordinate.

Due to the balance equation for the internal energy (31), with $r=0$, and deriving Eq. (59) with respect to the space, straightforward calculations show that

$$\ddot{e}+\frac{1}{\tau}\dot{e}-\frac{1}{\tau}\frac{\kappa}{c_v}e_{,xx}+\frac{f_1}{\kappa}q_{x_0}\dot{e}_{,x}=0. \tag{60}$$

Hyperbolic Eq. (60) governs the propagation of the pulses along the heat flux in a rigid heat conducting body. It is more general than the *telegraph equation* (28) derived in Sec. II, and it leads to the results

$$U_+=U_0(\sqrt{\phi^2+1}-\phi), \tag{61}$$

for the speeds of propagation of the pulses in the positive direction, namely, in the same direction as the heat flux, and

$$U_-=U_0(\sqrt{\phi^2+1}+\phi), \tag{62}$$

in the opposite direction, with $U_0=\sqrt{\frac{\kappa}{\tau c_v}}$ as the speed of propagation of thermal pulses in an equilibrium reference state, and $\phi=\frac{1}{2}\frac{f_1}{\kappa}\frac{q_{x_0}}{U_0}$.

From Eqs. (61) and (62) it follows that a small heat pulse will travel with different velocity in the direction of the heat flow than in the opposite direction. Taking into account Eq. (55), one may obtain

$$\Delta U=U_--U_+=2U_0\phi=\frac{1}{s_\beta}\frac{\partial s_\beta}{\partial e}q_{x_0}. \tag{63}$$

It is worth observing that the sign of ΔU is strictly connected with that of $\frac{1}{s_\beta}\frac{\partial s_\beta}{\partial e}$.

Equation (63) is a thermodynamic prediction of a relation between the speeds of thermal pulses in equilibrium, which give information on τ, and the speeds of thermal pulses under a heat flux. A positive value of ΔU is usual in the case of phonon hydrodynamics.[35]

Now we want to compare Eq. (63) with two other approaches, which have also dealt with second-sound propagation in the presence of heat flux. Let us start with the approach proposed by Coleman *et al.*[43] in 1982, which also describes a different speed of propagation toward or backward an average nonvanishing heat flux. In Ref. 43 a thermodynamic state space spanned by the absolute temperature θ and the heat flux q_i is supposed. If one neglects the terms quadratic with respect to the heat flux, for the difference of both speeds of propagation $\Delta U=U_--U_+$, the following relation holds:

$$\Delta U=U_--U_+=\frac{2a(T)}{c_v z(T)}q_{x_0}, \tag{64}$$

with $z(T)=\frac{\tau(T)}{\kappa(T)}$ and $a(T)=-\frac{T^2}{2}\frac{\partial z}{\partial T}$. Experimental analysis of second sound in solids shows that $\frac{\partial z}{\partial T}<0$,[43] and thus, it follows that if $q_{x_0}>0$ in the direction of increasing x, then $|U_-|$ exceeds $|U_+|$. Thus, the heat pulse propagating in a body conducting heat will travel more slowly in the direction of heat flow than in the opposite direction.

In Ref. 44 the same problem was studied but in the framework of extended irreversible thermodynamics, i.e., starting from a thermodynamic state space spanned by the internal energy e and the heat flux q_i, and combining the energy balance law Eq. (31), the Maxwell-Cattaneo equation (3), and the generalized equation of state for the thermodynamic absolute temperature θ,[4,44]

$$\frac{1}{\theta}=\frac{1}{T}-\frac{1}{2}\frac{\partial}{\partial e}\left(\frac{\tau}{\kappa T^2}\right)q_i q_i. \tag{65}$$

That way, the following hyperbolic equation is recovered:

$$A\ddot{e}+B\dot{e}_{,x}-e_{,xx}=0, \tag{66}$$

with $A=\frac{c_v\tau}{\kappa}$ and $B=2A(\frac{\kappa T^2}{\tau})\frac{\partial}{\partial e}(\frac{\tau}{\kappa T^2})q_{x_0}$.

Equation (66) leads ΔU to the result

$$\Delta U=U_- - U_+ = -\frac{\kappa T^2}{\tau}\frac{\partial}{\partial e}\left(\frac{\tau}{\kappa T^2}\right)q_{x_0}, \tag{67}$$

and so a wave traveling more slowly in the same direction of heat pulse than that in the opposite direction is recovered provided that $\frac{\partial}{\partial e}(\frac{\tau}{\kappa T^2})<0$. In Refs. 4 and 44 it is shown in a microscopic model for a phonon gas that this inequality is satisfied. It is easy to see that Eqs. (64) and (67) are equivalent, and they are also equivalent with Eq. (63) if s_β is identified as $s_\beta=\frac{\tau\kappa}{T^2}$. The thermodynamic consistency of this last assumption will be shown in Sec. VI A, where our approach will be compared with that of extended irreversible thermodynamics.[1,4] Thus, it is interesting to see that this phenomenology would have been lost if the usual Liu technique had been used without the extensions included in Eq. (40).

V. ENTROPY AND NONLINEAR HEAT TRANSPORT

The requirement that entropy is maximum at equilibrium stable states imposes some restrictions on the equations of state, which are useful to predict, for instance, the conditions for phase transitions. Instead, out of equilibrium the generalized entropy provides another kind of restrictions, related to the hyperbolic character of the evolution equations, and which are also related to the second differential of the entropy. Thus, the aims of the present section are both inferring the compatibility conditions of Eq. (52) with the principle of maximum entropy at equilibrium states and investigating its compatibility with the condition of convexity of the entropy function in nonequilibrium steady states.[4,5,18]

A. Equilibrium and steady states

For the sake of simplicity, we restrict ourselves to the problem of heat transport in a one-dimensional rigid body. At equilibrium states, we postulate the caloric equation of state

$$e=H(\beta), \tag{68}$$

with H as a regular function.

In Sec. IV A we have obtained that the entropy should be $s=s(e;\beta;\beta_{,k})$, and from Eq. (52) the matrix of the second-order derivatives $\delta^2 s$ may be written as

$$\delta^2 s=\left\|\begin{matrix} \frac{\partial^2 s_0}{\partial e^2}-\frac{1}{2}\frac{\partial^2 s_\beta}{\partial e^2}\beta_{,x}^2 & \frac{\partial^2 s_0}{\partial e\,\partial\beta}-\frac{1}{2}\frac{\partial^2 s_\beta}{\partial e\,\partial\beta}\beta_{,x}^2 & -\frac{\partial s_\beta}{\partial e}\beta_{,x} \\ \frac{\partial^2 s_0}{\partial\beta\,\partial e}-\frac{1}{2}\frac{\partial^2 s_\beta}{\partial\beta\,\partial e}\beta_{,x}^2 & \frac{\partial^2 s_0}{\partial\beta^2}-\frac{1}{2}\frac{\partial^2 s_\beta}{\partial\beta^2}\beta_{,x}^2 & -\frac{\partial s_\beta}{\partial\beta}\beta_{,x} \\ -\frac{\partial s_\beta}{\partial e}\beta_{,x} & -\frac{\partial s_\beta}{\partial\beta}\beta_{,x} & -s_\beta \end{matrix}\right\|, \tag{69}$$

where x is the linear spatial coordinate. At the equilibrium state, where $\beta_{,x}=0$, matrix (69) yields

$$\delta^2 s|_{\text{eq}}=\left\|\begin{matrix} -\frac{1}{c_v}\left(\frac{1}{T}\right)^2 & -\frac{1}{c_v}\frac{dH}{d\beta}\left(\frac{1}{T}\right)^2 & 0 \\ -\frac{1}{c_v}\frac{dH}{d\beta}\left(\frac{1}{T}\right)^2 & -\frac{1}{c_v}\left(\frac{1}{T}\frac{dH}{d\beta}\right)^2+\frac{1}{T}\frac{d^2H}{d\beta^2} & 0 \\ 0 & 0 & -s_\beta|_{\text{eq}} \end{matrix}\right\|. \tag{70}$$

The requirement that the entropy is maximum at equilibrium (for isolated systems) implies

$$c_v>0, \tag{71}$$

$$s_\beta|_{\text{eq}}>0, \tag{72}$$

$$\frac{d^2H}{d\beta^2}<0, \tag{73}$$

$$\frac{1}{c_v}\frac{1}{\theta^3}\frac{d^2H}{d\beta^2}\le 0. \tag{74}$$

It is easily seen that Eq. (74) is automatically fulfilled if relations (71) and (73) hold. Thus, for a one-dimensional rigid body conditions (71)–(73) are sufficient to have a maximum of entropy at equilibrium for hyperbolic heat transport, provided that Eq. (52) is postulated.

Far-from equilibrium, simple calculations lead from Eq. (69) to

$$\delta^2 s = \left\| \begin{matrix} -\frac{1}{c_v}\left(\frac{1}{\theta}\right)^2 & \frac{1}{\beta}\frac{\partial\gamma}{\partial e} & -\frac{\partial\theta}{\partial\beta_{,x}}\left(\frac{1}{\theta}\right)^2 \\ \frac{1}{\beta}\frac{\partial\gamma}{\partial e} & -\frac{\gamma}{\beta^2}+\frac{1}{\beta}\frac{\partial\gamma}{\partial\beta} & \frac{1}{\beta}\frac{\partial\gamma}{\partial\beta_{,x}} \\ -\frac{\partial\theta}{\partial\beta_{,x}}\left(\frac{1}{\theta}\right)^2 & \frac{1}{\beta}\frac{\partial\gamma}{\partial\beta_{,x}} & -s_\beta \end{matrix} \right\|, \tag{75}$$

where γ denotes a regular function such that

$$\frac{\partial s}{\partial\beta} = \frac{1}{\beta}\gamma(e;\beta;\beta_{,x}). \tag{76}$$

By design, γ at equilibrium tends to $\frac{c_v}{T}F(T)(\frac{dF}{dT})^{-1}$, where $F(T)=\beta|_{\text{eq}}$. Furthermore, we suppose that the derivatives of γ with respect to the state variables are of the first order of magnitude, and hence, approaching the stability of entropy far-from equilibrium, all the terms which contain products of these quantities will be neglected. Let us note that in matrix (75), both γ and θ depend on $\beta_{,x}$.

The requirement that the entropy is a convex function implies that matrix (75) must be negative definite. This yields

$$c_v > 0, \tag{77}$$

$$s_\beta(e;\beta) > 0 \quad \forall\,(e;\beta), \tag{78}$$

$$\frac{\partial\gamma}{\partial\beta} < \frac{\gamma}{\beta} \quad \forall\,(e;\beta;\beta_{,x}), \tag{79}$$

$$-\frac{1}{c_v}\left(\frac{1}{\theta}\right)^2 < 0 \quad \forall\,(e;\beta;\beta_{,x}), \tag{80}$$

$$-\frac{s_\beta}{c_v}\left(\frac{\gamma}{\beta}-\frac{\partial\gamma}{\partial\beta}\right) < 0 \quad \forall\,(e;\beta;\beta_{,x}). \tag{81}$$

Again, it is not difficult to verify that conditions (80) and (81) are automatically satisfied if Eqs. (77)–(79) are true. Thus, we may conclude that conditions (77)–(79) are sufficient to ensure that the entropy given by Eq. (52) is a convex function.

B. Nonlinear thermal conductivity

As previously said, in matrix (75) the functions θ and γ depend on $\beta_{,x}$, and therefore it incorporates an implicit influence of the semiempirical temperature gradient. To make this influence explicit, we will illustrate the analysis in the particular situation $s=s(e;\beta_{,x})$. In this case, Eq. (69) reduces to

$$\delta^2 s = \left\| \begin{matrix} -\frac{1}{c_v}\left(\frac{1}{T}\right)^2 - \frac{1}{2}\frac{\partial^2 s_\beta}{\partial e^2}\beta_{,x}^2 & -\frac{\partial s_\beta}{\partial e}\beta_{,x} \\ -\frac{\partial s_\beta}{\partial e}\beta_{,x} & -s_\beta \end{matrix} \right\|. \tag{82}$$

Two immediate stability conditions will be $c_v>0$ and $s_\beta(e)>0$, which are the same as in Eqs. (77) and (78). Another condition, which explicitly exhibits the nonequilibrium influence through the nonequilibrium parameter $\beta_{,x}$, refers to the positive sign of the complete determinant and is

$$\frac{1}{c_v}\left(\frac{1}{T}\right)^2 + \frac{1}{2}\frac{\partial^2 s_\beta}{\partial e^2}\beta_{,x}^2 > 0, \tag{83}$$

where every derivatives of the function $s_\beta(e)$ with respect to the state variables are supposed to be of the first order of magnitude. Relation (83) will be fulfilled if

$$-\beta_{,x}^2 < \frac{2}{c_v}\left(\frac{1}{T}\right)^2\left(\frac{\partial^2 s_\beta}{\partial e^2}\right)^{-1}, \tag{84}$$

which represents a constraint on the admissible values of the heat flux. A similar result has been obtained in Ref. 25. It is worth observing that function κ appearing in Eq. (1) cannot be identified with the classical Fourier heat conductivity, although it should reduce to such a function when β coincides with T. In order to point out the relation between these two material functions, let us define an effective conductivity κ_{eff} as

$$q_x = -\kappa_{\text{eff}}\theta_{,x}, \tag{85}$$

where θ denotes the nonequilibrium absolute temperature. Due to the constitutive Eq. (36), κ_{eff} contains dynamical terms which are capable to reproduce the hyperbolic regime. The classical parabolic behavior is recovered when the nonequilibrium temperature θ reduces to the equilibrium temperature T. In such a case κ_{eff} reduces to Fourier's heat conductivity.

On the other hand, following the ideas of Onsager's linear thermodynamics,[42] let us write

$$\begin{aligned} q_x &= \Gamma\left(\frac{\partial s}{\partial e}\right)_{,x} \\ &= \Gamma\left[\frac{1}{T} - \frac{1}{2}\frac{\partial s_\beta}{\partial e}\beta_{,x}^2\right]_{,x} = -\frac{\Gamma}{T^2}\left[1 + T^2\frac{c_v}{2}\frac{\partial^2 s_\beta}{\partial e^2}\beta_{,x}^2\right]\theta_{,x}, \end{aligned} \tag{86}$$

with Γ as a positive phenomenological coefficient related to the thermal conductivity. In deriving Eq. (86) we have neglected the contribution of the second spatial derivatives of β, which are related to the first spatial derivatives of the heat flux, since we are looking for a form of Eq. (85) which is capable to reproduce the hyperbolic regime, namely, a Maxwell-Cattaneo-type equation. This is not guaranteed if the first spatial derivatives of the heat flux $\mathbf{q}$ enter its evolution equation. The entropy rate of change in this case is

$$\dot{s} = \left(\frac{\partial s}{\partial e}\right)\dot{e} = -\left(\frac{\partial s}{\partial e}q_x\right)_{,x} + q_x\left(\frac{\partial s}{\partial e}\right)_{,x}. \tag{87}$$

The first term in the right-hand side of Eq. (87) is the divergence of the entropy flux and the second one the entropy production, which, in view of Eq. (86), is simply $\sigma^{(\beta)} = \Gamma(\frac{\partial s}{\partial e})_{,x}(\frac{\partial s}{\partial e})_{,x}$. At the equilibrium, where $\frac{\partial s}{\partial e}=\frac{1}{T}$, relation (86) is simply the Fourier law, provided that the heat conductivity is given by $\frac{\Gamma}{T^2}$. From Eq. (86), supposing $\kappa=\frac{\Gamma}{T^2}$, we obtain an effective nonlinear thermal conductivity which depends on $\beta_{,x}$,

$$\kappa_{\text{eff}} = \kappa\left[1 + T^2\frac{c_v}{2}\frac{\partial^2 s_\beta}{\partial e^2}\beta_{,x}^2\right]. \tag{88}$$

When $\frac{\partial^2 s_\beta}{\partial e^2}<0$, relation (88) implies a reduction in thermal conductivity for higher temperature gradients. Such a reduction is indeed observed in far-from equilibrium situations.[45–50] However, for arbitrary values of $\beta_{,x}$, the decrease in Eq. (88) would be too drastic, as it could lead to a negative value for the effective thermal conductivity. This would imply that internal energy tends to accumulate in a point rather than being distributed in a homogeneous way. However, if $\frac{\partial^2 s_\beta}{\partial e^2}<0$, i.e., $\kappa_{\text{eff}}<\kappa$, inequality (84) ensures the positiveness of κ_{eff}. If, instead, $\frac{\partial^2 s_\beta}{\partial e^2}>0$, then $\kappa_{\text{eff}}>\kappa$. Let us note that in this case, Eq. (83) is fulfilled whatever the value of $\beta_{,x}$ is, namely, there is no constraint on the admissible values of the heat flux. Moreover, it follows from Eq. (88) that κ_{eff} reduces to κ at the equilibrium where $\beta_{,x}=0$. Finally, both κ and κ_{eff} yield the Fourier heat conductivity if $\beta\equiv\theta\equiv T$.

Conductivity (88) is specially interesting from the perspective of nonlinear heat transport, as it depends on $\beta_{,x}^2$, which is a nonlinear and nonlocal term. A nonlocal and nonlinear heat conductivity arises in several situations involving finite speed of propagation of thermal disturbances, such as in radiative heat transfer or in plasma physics. In these phenomena the heat flux cannot assume arbitrary values, but it is bounded by a saturation value, of the order of the energy times the maximum speed of propagation. The saturation effect implies a drastic reduction in the flux with respect to the values predicted by Fourier law and can be interpreted by introducing an effective thermal conductivity, depending nonlinearly on the temperature gradient too. A typical form of the heat flux is[45–50]

$$q_x = -\frac{\kappa}{\sqrt{1+\frac{al^2}{\theta^2}\theta_{,x}^2}}\theta_{,x}, \tag{89}$$

with l as the mean-free path and a is a numerical dimensionless parameter.

In a small system of size L and submitted to a difference in temperature $\Delta\theta$, the temperature gradient will be of the order of $\frac{\Delta\theta}{L}$, and the second term under the square root in the denominator of Eq. (89) will be of the order of $a(\frac{l}{L})^2(\frac{\Delta\theta}{\theta})^2$. For $L\gg l$, Eq. (89) reduces to Fourier law. In the ballistic limit when $l\gg L$ and recalling that the thermal conductivity is $\kappa=\frac{1}{3}\varrho c_v l\bar{v}$, with $\bar{v}$ as the average value of the modulus of the particle speeds, Eq. (89) yields for the modulus of the heat flux $q_x=\frac{1}{3\sqrt{a}}\varrho c_v\theta\bar{v}$. This corresponds to the ballistic transport of heat flux for high values of the gradient. Indeed, the maximum value of $\frac{\Delta\theta}{L\theta}$, with $\Delta\theta=\theta_2-\theta_1$ and $\theta=\frac{\theta_2+\theta_1}{2}$, will be achieved when L is small and when $\theta_2\gg\theta_1$ or $\theta_1\gg\theta_2$, in which case the average θ will be of the order of the increment $\Delta\theta$. It is logical to expect that for a given average temperature, the maximum value of the heat flux will not be arbitrarily high but bounded by the ballistic value in which most of the particles cross the system without experiencing collisions with other particles.

The presence of the mean-free path and the relevance of the denominator in ballistic situations, which we have just commented explicitly, outline the expected importance of this nonlinear reduction in the thermal conductivity in nanosystems smaller than the mean-free path.

In terms of β, the effective thermal conductivity appearing in the equation above can be expressed as

$$\kappa_{\text{eff}} = \frac{\kappa}{\sqrt{1+\frac{al^2}{\beta^2}\beta_{,x}^2}}. \tag{90}$$

This describes a nonlinear reduction in thermal conductivity which does not become negative for any value of $\beta_{,x}$. If we perform an expansion of Eq. (90) around the equilibrium, (i.e., $\beta_{,x}=0$), we see that Eq. (88) represents a second-order approximation of Eq. (90) provided that

$$T^2 c_v\frac{\partial^2 s_\beta}{\partial e^2} = -\frac{al^2}{\beta^2}. \tag{91}$$

VI. COMPARISON WITH EIT APPROACH AND WITH THE GENERALIZED ONSAGER FORMALISM

In this section we will compare our approach to two other thermodynamic approaches: extended irreversible thermodynamics (EIT), incorporating nonlocal effects, and a generalization of the Onsager version of nonequilibrium thermodynamics.

In EIT,[1,4,5] the fluxes (heat flux and higher order fluxes) are used as independent variables, instead of ∇e and $\nabla\beta$, and they lead to relaxational equations with nonlinear contributions which generalize the Maxwell-Cattaneo Eq. (3). Concerning Onsager's approach, it is in some aspects the opposite way that we have explored: instead of starting from some evolution equations and searching for the form of the generalized entropy, it assumes a generalized entropy and looks for the corresponding evolution equations satisfying the requirement of a positive-definite entropy production.

A. EIT approach

EIT is a nonequilibrium thermodynamic theory where the fluxes, rather than the gradients, are used as independent variables to include nonlocal effects.[1,4] In order to describe nonlocal effects, in the framework of EIT it is assumed that the entropy depends on the energy and the heat flux, and it is shown that the generalized entropy compatible with the Maxwell-Cattaneo equation has the form[4,51]

$$s(e;q_i) = s_{\text{eq}}(e) - \frac{\tau}{2\kappa T^2}q_i q_i. \tag{92}$$

If the constitutive assumption (36) holds, then Eq. (92) may be rewritten as $s(e;\beta_{,i})=s_{\text{eq}}(e)-\frac{\tau\kappa}{2T^2}\beta_{,i}\beta_{,i}$ and it allows to interpret the s_β coefficient appearing in Eq. (52) as

$$s_\beta(e;\beta) = \frac{\tau\kappa}{T^2}. \tag{93}$$

Let us point out that due to Eq. (93), from Eq. (63) it follows that a small heat pulse propagating in a rigid heat conducting body will travel more slowly in the direction of the heat flow than in the opposite direction according to Refs. 43 and 44 as can been seen in Sec. IV B.

Furthermore, in EIT the relation between the thermodynamic absolute temperature θ, obtained from the relation $\theta^{-1} = (\frac{\partial s}{\partial e})$, and the local-equilibrium one T, defined in terms of the internal energy density, without reference to entropy, is given by

$$\theta = T + \frac{T^2}{2}\frac{\tau}{\kappa}\frac{\partial}{\partial e}\left(\frac{1}{T^2}\right)q_i q_i \tag{94}$$

if the relaxation time τ and the effective thermal conductivity κ are constant.[1,4]

Let us observe that Eq. (94) is in accordance with the generalized equation of state for the thermodynamic absolute temperature (65). Indeed, from Eq. (65) it follows that

$$\theta = T\left[1 - \frac{T}{2}\frac{\partial}{\partial e}\left(\frac{\tau}{\kappa T^2}\right)q_i q_i\right]^{-1}. \tag{95}$$

Thus, performing an expansion of Eq. (95) around the equilibrium state (i.e., $q_i = 0$), up to the first-order one has

$$\theta = T\left[1 + \frac{T}{2}\frac{\partial}{\partial e}\left(\frac{\tau}{\kappa T^2}\right)q_i q_i\right], \tag{96}$$

and Eq. (94) may be recovered.

On the other hand, from Eq. (32), due to relations (54)–(56) and the constitutive assumption (36) we have $\dot{\beta} = -\frac{1}{\tau}(\beta - \theta) + \frac{1}{2}f_1\beta_{,i}\beta_{,i}$, and it follows that in the steady state, i.e., $\dot{\beta} = 0$, the semiempirical temperature β will not become equal to θ but we will have, recalling Eq. (55),

$$\beta = \theta + \frac{\tau}{2\kappa}\frac{1}{s_\beta}\frac{\partial s_\beta}{\partial e}q_i q_i. \tag{97}$$

It is easy to observe that, taking into account Eq. (93), relation (97) reduces to Eq. (94) once one takes into consideration the different notations in Eqs. (94) and (97), namely, if one identifies T with θ and θ with β. Thus, the difference between β and θ gives another analogy between our formalism and EIT. Note again that the fact that $f_1 \neq 0$ is essential for this analogy. However, in the present formalism the dynamical character of temperature plays a central role, which is not so explicitly seen in EIT. Thus, the combination of both theories may give a deeper insight into temperature.

B. Onsager's formalism

We will now take the generalized entropy (52) as a starting hypothesis, and we will look for the corresponding evolution equation following from it in the framework of the Onsagerian classical irreversible thermodynamics.[21] By focusing our attention on hyperbolic heat transport, we will suppose $s = s(e;\beta_{,i})$. Thus, taking into account Eqs. (35) and (53), from the extended Gibbs equation we get

$$\dot{s} = \frac{1}{\theta}\dot{e} - s_\beta\beta_{,i}\dot{\beta}_{,i}. \tag{98}$$

If $r = 0$, due to the local balance of internal energy (31), from Eq. (98) it follows that

$$\dot{s} + \left(\frac{1}{\theta}q_i\right)_{,i} = -\frac{1}{\theta^2}q_i\theta_{,i} - s_\beta\beta_{,i}\dot{\beta}_{,i}. \tag{99}$$

According to Onsager theory,[19–21] we take q_i and $\dot{\beta}_{,i}$ as fluxes and $-\frac{1}{\theta^2}\theta_{,i}$ and $-s_\beta\beta_{,i}$ as thermodynamic forces. Expressing the former one as linear combination of the latter, we have

$$q_i = -\frac{L_{11}}{\theta^2}\theta_{,i} - L_{12}s_\beta\beta_{,i}, \tag{100}$$

$$\dot{\beta}_{,i} = -\frac{L_{21}}{\theta^2}\theta_{,i} - L_{22}s_\beta\beta_{,i}, \tag{101}$$

where L_{11}, L_{12}, L_{21}, and L_{22} are the phenomenological coefficients. In fact, Eq. (101) may be seen as the gradient extension of the dynamical Eq. (2) for β, if the term $L_{22}s_\beta$ is interpreted as $\frac{1}{\tau}$. The positiveness of entropy production requires that

$$L_{11} > 0, \tag{102}$$

$$L_{22} > 0, \tag{103}$$

$$L_{11}L_{22} - L_{12}L_{21} \geq 0. \tag{104}$$

Differentiating expression (100) with respect to time, in the case of constant phenomenological coefficients, straightforward calculations yield

$$\dot{q}_i + L_{22}s_\beta q_i = -\frac{s_\beta}{\theta^2}(L_{11}L_{22} - L_{12}L_{21})\theta_{,i} - \frac{L_{11}}{\theta^2}\dot{\theta}_{,i}, \tag{105}$$

which is the double-lag heat transfer equation,[11] exhibiting relaxational effects both in the heat flux and in the temperature gradient.

From Onsager-Casimir point of view, the analysis of Eq. (105) depends on the time-reversal parity assigned to $\dot{\beta}$. If it is assumed an odd variable with respect to time reversal, as q_i, then $L_{12} = L_{21}$. But then, due to Eq. (104), L_{11} and L_{22} will be necessarily different from zero. This implies that in Eq. (105) the term $\frac{L_{11}}{\theta^2}\dot{\theta}_{,i}$ cannot be eliminated. However, due to the considerations of Sec. II, the Maxwell-Cattaneo equation can be recovered at a time scale at which L_{11} is negligible. In such a case, the semiempirical heat conduction law (1) follows from Eq. (100), and this proves that heat conduction theory developed in this paper is in accordance with Onsager's approach to nonequilibrium thermodynamics. Identification of the terms in Eq. (105) leads to

$$L_{22}s_\beta = \frac{1}{\tau}, \tag{106}$$

for the relaxation time and

$$\kappa = \frac{L_{11}L_{22} - L_{12}L_{21}}{\theta^2}, \tag{107}$$

for the thermal conductivity.

If one takes $L_{12}=L_{21}=0$, then Eq. (105) may be written as

$$\tau\dot{q}_i + q_i = -\kappa\theta_{,i} - \kappa\tau\dot{\theta}_{,i}, \tag{108}$$

where now $\kappa=\frac{L_{11}}{\theta^2}$. If $L_{12}=L_{21}\neq 0$, from Eq. (105) instead one has

$$\tau\dot{q}_i + q_i = -\kappa\theta_{,i} - \tau\kappa'\dot{\theta}_{,i}, \tag{109}$$

where the thermal conductivity is given by Eq. (107) and

$$\kappa' = \frac{L_{11}}{\theta^2}. \tag{110}$$

VII. CONCLUDING REMARKS

In this paper we have revisited the nonlocal and the nonlinear heat transport in a rigid body in terms of a dynamical semiempirical temperature β. Such an analysis is motivated by the increasing importance of the application of generalized heat transport equations in nanotechnology,[11,52–55] and its results may have special interest for heat transport in miniaturized systems with internal degrees of freedom or for the analysis of high-frequency short-wavelength perturbations. The result we have studied, concerning the different speed of propagation of thermal signals along or against the average heat flux, indicates that perturbations of the hotter temperature or of the lower temperature in a nanosystem will not propagate at the same speed inside the system. Furthermore, the understanding of temperature in nonequilibrium states is a fundamental topic and an active field of research,[16] and therefore the exploitation of the consequences of dynamical temperatures in heat transport may be useful also in this field.

Starting from a thermodynamic state space spanned by the specific internal energy e, the semiempirical temperature β, and their first gradients ∇e and $\nabla\beta$, in the case of Eq. (32), which leads to an hyperbolic regime by the extended Liu procedure,[8] we found that the entropy density is nonlocal only with respect to β, as given explicitly by Eq. (52).[56] In particular, in Sec. IV B some physical consequences of this contribution on the propagation of second sound in the presence of a heat flux have been pointed out.

We have emphasized the importance of the generalized version of the Liu procedure for the exploitation of the second law. Without this gradient extension, the effects explored in this paper would vanish. However, they are consistent with the result obtained, some years ago, from two different versions of extended thermodynamics. This agreement emphasizes the physical interest of the nonlocal and nonlinear terms considered in this paper.

The analysis of the hyperbolic regimes is based on purely thermodynamic grounds. Furthermore, in order to give a deeper insight to the dynamical semiempirical temperature and the subsequent implications for the form of the entropy and the transport equation for the heat flux, all the results have been compared both with EIT and with the Onsager formalism.

Finally, we feel that the analysis for nonlinear hyperbolic heat transfer is specially interesting, as some of the stability conditions may be related to nonlinear contributions to a generalized thermal conductivity. In the future, nonlinear effects should be paid more attention. For instance, another interesting aspect of propagation of heat pulses in the presence of a temperature gradient is related to the variation in the speed U as a function of the temperature. From experimental observation in crystals of NaF and of Bi at low temperatures,[26,43,57] it follows that $\frac{\partial U}{\partial\theta}<0$. This implies that if a rectangular-shaped heat pulse propagates in the same direction of the temperature gradient, namely, in the opposite direction of heat flux, the pulse becomes narrower and higher because the frontal edge, being always at higher temperature than the rear edge, has a lower velocity than the rear edge, and the distance between them slowly decreases. This process cannot last indefinitely, as it would lead to a singularity. Thus, nonlinear effects in the propagation velocity would have an increasing importance, and they should deserve more attention in the future.

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