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Nonequilibrium temperatures, heat waves, and nonlinear heat transport equations

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A dynamical nonequilibrium temperature has been proposed to describe relaxational equations for the heat flux. This temperature provides an alternative description to the Maxwell-Cattaneo equation. In the linear regime and in bulk systems both descriptions are equivalent but this is not so when nonlinear effects are included. Here we explore the influence of nonlinear terms on the phase speed of heat waves in nonequilibrium steady states in both theoretical models and we show that their predictions are different. This could allow to explore which description is more suitable, when experiments on these situations will become available. Furthermore, we have analyzed a nonlinear and nonlocal constitutive equation for the heat flux and we have shown its analogy with the Navier-Stokes equation in the regime of phonon hydrodynamics in nanosystems. This analogy allows one to define a dimensionless number for heat flow, analogous to the Reynolds number, and to predict a critical heat flux where nonlinear effects could become dominant.

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I. INTRODUCTION

In classical thermodynamics the components of the heat flux **q** are given by the Fourier law in terms of the gradient of the local-equilibrium temperature T and the thermal conductivity. To describe the evolution of the temperature this law is combined with the energy-conservation equation. The results fit correctly in systems where the mean-free path of the heat carriers is much shorter than the characteristic size of the system and when the rate of variation in the heat flux is small enough, namely, when the frequency of the perturbations is sufficiently smaller than the reciprocal of the heat-relaxation time. Nevertheless, nowadays nanowires and thin layers are receiving much attention because of their potential applications in miniaturized devices. Heat transport in these devices differs significantly from the predictions of Fourier law[1–4] because their behavior is strongly influenced by nonlocal and nonlinear effects,[5,6] which lack in the Fourier law. Indeed, even a small difference in temperature, or electrical potential, over a small scale length may generate very high gradients.[7–9] Furthermore, in microdevices working at high frequencies, one has also to take into account the relaxation time of the heat flux[10] because heat flux will not have time enough to accommodate to the value given by the Fourier law. As a consequence, a more general heat-transport equation than the Fourier law must be introduced.[11–13]

Here we compare two descriptions of relaxational heat transfer. In one of them this is done in terms of a dynamical semiempirical temperature β.[14–16] Its essential idea is to assume that the heat flux q_i is given by

$$q_i = -\lambda \beta_{,i} \tag{1}$$

with λ as the thermal conductivity, and $\beta_{,i} \equiv \partial\beta/\partial x_i$ as the spatial derivative of β with respect to the Cartesian coordinate x_i. This allows us to generalize the evolution equation for the temperature in order to encompass nonlinear and relaxation effects. Equation (1) is capable to reproduce different heat-conduction regimes, depending on the evolution equation of β, whose simplest form is

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

where θ is the nonequilibrium absolute temperature, and τ_R is a relaxation time related, for instance, to resistive processes of interaction among the phonons, representing the heat carriers. In the hypothesis that the material functions τ_R and λ are constant, the combination of Eqs. (1) and (2) leads to the Maxwell-Cattaneo equation[14,17]

$$\tau_R \dot{q}_i + q_i = -\lambda \theta_{,i}, \tag{3}$$

which provides a well-known description of high-frequency thermal waves.

The second description is based on the Maxwell-Cattaneo equation [Eq. (3)] without using the concept of the dynamical temperature β, i.e., it starts directly from Eq. (3), without using Eqs. (1) and (2). The thermodynamical consistency of both models has been tested in several papers (see, for instance, Refs. 15 and 16 for the first model and Ref. 11 for the second model).

Nevertheless, Eqs. (2) and (3) do not describe nonlinear effects since they are obtained under the hypothesis of constant material functions. Nonlinear effects may be understood in two different ways: as a temperature dependence in the material functions or as the presence of nonlinear products of the temperature gradient (or the heat flux) in the transport equations. We will examine both kinds of nonlinearities.

In this paper we also explore the differences between the description based on the dynamical semiempirical temperature β through Eqs. (1) and (2) and the description based directly on Eq. (3), i.e., based on the nonequilibrium temperature θ and the Maxwell-Cattaneo equation. It is worth observing that in linear situations, namely, for constant values of the heat conductivity λ, the specific heat per unit volume c_v, and the relaxation time τ_R, Eqs. (1) and (2) are equivalent to Eq. (3). Hence, we restrict ourselves to nonlinear situations, to explore how the predictions of both models differ from each other. In particular, we will focus our attention on the influence of nonlinear effects on the phase speed of heat waves propagating along a nonequilibrium steady state. First of all, as heat-conduction measurements in crystals show clearly, the material functions λ, c_v, and τ_R are temperature dependent,[18,19] and such a dependency introduces nonlinearities in the evolution equation for the temperature. This kind of nonlinear effects, up to the second-order approximation, will be studied in Sec. II. Moreover, due to the small sizes of the systems, the temperature gradient and heat flux may reach very high values. Therefore the nonlinearity arising from second-order products of these quantities, implying a nonlinear dependence of the transport law on the temperature gradient, as proposed in Eq. (27), will be considered in Sec. III. In Sec. IV we will combine nonlinear terms with nonlocal terms, as proposed in Eq. (43), and study their influence on the stability of the heat flux. In Sec. V we investigate the compatibility of the approximation up to the second order of the evolution equations with the entropy principle. More precisely, we show that, under the hypothesis that third-order quantities are negligible, both the models considered here still satisfy second law of thermodynamics. The section contains also a table with the main results on second-sound propagation.

II. HEAT-WAVE PROPAGATION IN TEMPERATURE-DEPENDENT MATERIAL FUNCTIONS

As said above, for constant material functions, hypotheses in Eqs. (1) and (2) and that in Eq. (3) are equivalent in the sense that the assumption of a heat flux related to the dynamical temperature β, obeying the relaxational Eq. (2) gives for the temperature the same results as a heat flux related to the nonequilibrium temperature θ through Eq. (3). This is not necessarily true if the material functions are no longer supposed to be constant. Thus, we remove here this hypothesis and suppose that the following assumptions:

$$c_v = c_v(\theta), \quad \tau_R = \tau_R(\theta), \quad \lambda = \lambda(\theta) \tag{4}$$

hold for the material functions. Then, once one has taken into account Eq. (2) and the constitutive assumption in Eq. (1), straightforward calculations point out that the evolution equation for the heat flux is given by

$$\tau_R \dot{q}_i + q_i\left(1 - \frac{\tau_R}{\lambda}\frac{\partial \lambda}{\partial \theta}\dot{\theta}\right) = -\lambda\left(1 - \frac{\partial \tau_R}{\partial \theta}\dot{\beta}\right)\theta_{,i}, \tag{5}$$

instead of the Maxwell-Cattaneo equation [Eq. (3)]. Then, the combination of Eq. (5) with the local balance of specific internal energy e, namely,

$$\dot{e} = -q_{i_{,i}}, \tag{6}$$

yields the following hyperbolic equation for the nonequilibrium temperature θ:

$$\begin{aligned}\tau_R c_v \ddot{\theta} + c_v \dot{\theta} + \tau_R\left(\frac{\partial c_v}{\partial \theta} - \frac{c_v}{\lambda}\frac{\partial \lambda}{\partial \theta}\right)\dot{\theta}^2 &= \lambda\left(1 - \frac{\partial \tau_R}{\partial \theta}\dot{\beta}\right)\theta_{,ii} - \frac{\tau_R}{\lambda}\frac{\partial \lambda}{\partial \theta}q_i\dot{\theta}_{,i} \\ &\quad - \left[\left(\frac{2\lambda}{\tau_R}\frac{\partial \tau_R}{\partial \theta} - \frac{\partial \lambda}{\partial \theta}\right)\theta_{,i} + \frac{2}{\tau_R}\frac{\partial \tau_R}{\partial \theta}q_i\right]\theta_{,i},\end{aligned} \tag{7}$$

wherein for the sake of simplicity we have neglected the third-order terms, as in what follows. To be more explicit, we consider each time and/or space derivative as a first-order term. That way, nonlinear terms in $\dot{\theta}\theta_{,i}q_i$ or in $\dot{\beta}\theta^2_{,i}$ have been neglected. In Sec. V we comment about this approximation more deeply.

In order to show the consequences of the constitutive assumptions in Eq. (4), let us consider now the propagation of longitudinal plane temperature waves

$$\theta(x;t) = \bar{\theta}(x) + \theta_0 \exp[i(\omega t - kx)], \tag{8}$$

which may be experimentally realized by imposing at one end of the system a sinusoidally time-dependent temperature perturbation from a stationary reference level $\bar{\theta}(x)$ and studying the behavior of the temperature perturbation at different points along the system. Then, limiting ourselves to the one-dimensional case, in a linearized approach around a nonequilibrium steady state with an average heat flux $q_{x_0} = -\lambda\bar{\theta}_{,x}$, Eq. (7) leads to

$$\tau_R c_v \ddot{\theta} + c_v \dot{\theta} = \lambda \theta_{,xx} - \frac{\partial \ln \lambda}{\partial \theta}q_{x_0}\theta_{,x} - \tau_R\frac{\partial \ln \lambda}{\partial \theta}q_{x_0}\dot{\theta}_{,x}, \tag{9}$$

being x the longitudinal spatial coordinate. It is useful to underline that in the right-hand side of Eq. (9) the two quantities $(\partial \ln \lambda / \partial \theta)q_{x_0}$ and $\tau_R(\partial \ln \lambda / \partial \theta)q_{x_0}$ are taken as fixed parameters. Moreover, nonlinear second-order terms, as, for instance, $\dot{\theta}^2$ and $\theta^2_{,x}$ (related to the perturbation in θ), have been neglected because we will only consider small-amplitude waves.

From the other hand, the combination of Eqs. (3) and (6) yields

$$\begin{aligned}\tau_R c_v \ddot{\theta} + c_v \dot{\theta} + \tau_R\frac{\partial c_v}{\partial \theta}\dot{\theta}^2 &= \lambda \theta_{,ii} - \left[\left(\frac{\lambda}{\tau_R}\frac{\partial \tau_R}{\partial \theta} - \frac{\partial \lambda}{\partial \theta}\right)\theta_{,i} + \frac{1}{\tau_R}\frac{\partial \tau_R}{\partial \theta}q_i\right]\theta_{,i},\end{aligned} \tag{10}$$

whose linearization, in the one-dimensional case and around a nonequilibrium steady state with an average heat flux q_{x_0}, instead, produces

$$\tau_R c_v \ddot{\theta} + c_v \dot{\theta} = \lambda \theta_{,xx} - \frac{\partial \ln \lambda}{\partial \theta}q_{x_0}\theta_{,x}, \tag{11}$$

and thus, we are allowed to conclude that, in the aforementioned approximation, the models (5) and (3), leading, respectively, to Eqs. (9) and (11), yield different predictions arising from the temperature dependence of the thermal conductivity $\lambda(\theta)$.

In the case of high-frequency thermal waves (i.e., when $\dot{\theta} \ll \ddot{\theta}$ and $\theta_{,x} \ll \theta_{,xx}$), taking into account Eq. (8), from Eqs. (9) and (11), respectively, the following dispersion relations may be derived:

$$k^2 + \frac{\tau_R}{\lambda}\frac{\partial \ln \lambda}{\partial \theta} q_{x_0} \omega k - \frac{\tau_R}{\chi}\omega^2 = 0, \tag{12a}$$

$$k^2 - \frac{\tau_R}{\chi}\omega^2 = 0, \tag{12b}$$

where $\chi = \lambda / c_v$ is the thermal diffusivity.

From Eq. (12a), recalling that the phase velocity U is defined as $U \equiv |\omega / \mathrm{Re}(k)|$, we conclude that the model (5) predicts

$$U^{\pm} = \sqrt{\frac{\chi}{\tau_R}} \frac{1}{\sqrt{1+\Lambda^2} \pm \Lambda}, \tag{13}$$

where $2\Lambda = q_{x_0}(\sqrt{\tau_R / \lambda c_v})(\partial \ln \lambda / \partial \theta)$. The phase velocity U^+ corresponds to $k > 0$, namely, to a heat flux q_{x_0} flowing in the same direction of the longitudinal plane temperature waves [Eq. (8)] while the phase velocity U^-, corresponding to $k < 0$, holds in the opposite situation, i.e., for waves propagating in the opposite direction of q_{x_0}. When $\Lambda = 0$, i.e., if the heat conductivity λ does not depend on θ, from Eqs. (13) we have $U^+ \equiv U^- \equiv U_0 = \sqrt{\chi / \tau_R}$, which is the usual result for the speed of heat pulse or high-frequency heat waves, also predicted by Eq. (12b),[11–13,20] as it will be shown below.

Equations (13) point out that a small temperature pulse will travel with different velocity in the direction of the heat flow (U^+) than in the opposite direction (U^-). Similar results have been obtained in Refs. 14, 18, and 21. In particular, for $\Lambda \ll 1$ Eqs. (13) become

$$U^{\pm} = U_0(1 \mp \Lambda) \tag{14}$$

and the consequent difference in the phase velocities is

$$U^- - U^+ = \frac{q_{x_0}}{c_v}\frac{\partial \ln \lambda}{\partial \theta}. \tag{15}$$

To give an explicit estimation of the difference in Eq. (15), let us consider, for example, a specimen of NaF at low temperature. On the basis of the available experimental data,[22,23] for this system Cimmelli and Frischmuth[24] found that the specific heat c_v is given by

$$c_v(\theta) = (c_{v0} + c_{v1}\theta)\theta^3,$$

$$c_{v0} = 2.15 \ \mathrm{J\ cm^{-3}\ K^{-4}}, \quad c_{v1} = -0.02 \ \mathrm{J\ cm^{-3}\ K^{-5}}. \tag{16}$$

On the other hand, in the neighborhood of 15.05 K, namely, the value of the temperature at which the thermal conductivity of NaF attains the maximum ($\lambda = 240 \ \mathrm{W\ cm^{-1}\ K^{-1}}$),[22] the thermal conductivity is well represented by the function

$$\lambda(\theta) = e^{e^{\Lambda_1 + \Lambda_2 \ln \theta + \Lambda_3 \ln^2 \theta}},$$

$$\Lambda_1 = -7.15, \quad \Lambda_2 = 6.53, \quad \Lambda_3 = -1.20. \tag{17}$$

With these assumptions, the difference in Eq. (15) between the two phase velocities would be

$$U^- - U^+ = \frac{q_{x_0}(\Lambda_2 + 2\Lambda_3 \ln \theta)}{(c_{v0} + c_{v1}\theta)\theta^4} e^{\Lambda_1 + \Lambda_2 \ln \theta + \Lambda_3 \ln^2 \theta}. \tag{18}$$

For example, for a specimen of NaF at 15.05 K, with an average heat flux $q_{x_0} = 1.5 \times 10^{-4} \ \mathrm{W\ cm^{-2}}$ (corresponding to $\Lambda \approx 10^{-5}$ at 15.05 K), from Eq. (18) it follows that the difference in phase velocities is $U^- - U^+ = 2.1 \times 10^{-4} \ \mathrm{cm}\ \mu\mathrm{s}^{-1}$. It is worth noticing that the sign of $U^- - U^+$ may be different in other physical situations.

Equation (12b), instead, points out that the model (3) yields only one phase velocity for a high-frequency thermal wave, i.e.,

$$U^+ = U^- = U_0 = \sqrt{\frac{\chi}{\tau_R}}, \tag{19}$$

which will be also expressed in term of the phonons velocity $\bar{v}$. In fact, since for the thermal conductivity the relation $\lambda = c_v \tau_R \bar{v}^2 / 3$ holds, from Eq. (19) immediately follows $U_0 = \bar{v} / \sqrt{3}$. In particular, in the same temperature conditions as above, for a NaF specimen, the phase speed in Eq. (19) is $U_0 = 10.53 \ \mathrm{cm}\ \mu\mathrm{s}^{-1}$. Then, the value of the difference $U^- - U^+$, obtained from Eq. (18), is relatively small with respect to the value of U_0, but it has an interesting physical meaning, as we have shown.

In the opposite limit of low-frequency thermal-wave propagation, from Eq. (8) both models (3) and (5) yield the same dispersion relation, namely,

$$\omega c_v = k \frac{\partial \ln \lambda}{\partial \theta} q_{x_0} \tag{20}$$

and thus, in this case, it follows that the phase velocity is

$$U = \frac{q_{x_0}}{c_v}\frac{\partial \ln \lambda}{\partial \theta}. \tag{21}$$

In the general case of intermediate frequencies, from Eqs. (9) and (11) one may derive the following dispersion relations:

$$k^2 + \frac{q_{x_0}}{\lambda}\frac{\partial \ln \lambda}{\partial \theta}(\tau_R \omega - i)k - \frac{\omega}{\chi}(\tau_R \omega - i) = 0, \tag{22a}$$

$$k^2 - i\frac{q_{x_0}}{\lambda}\frac{\partial \ln \lambda}{\partial \theta} k - \frac{\omega}{\chi}(\tau_R \omega - i) = 0 \tag{22b}$$

and then, the consequent phase velocities

$$U^{\pm} = \frac{2}{\tau_R}\frac{\lambda}{q_{x_0}}\left[\frac{\partial \ln \lambda}{\partial \theta}\left(\sqrt[4]{A}\sqrt{\frac{1}{2} + \frac{A_1}{2\sqrt{A}}} \pm 1\right)\right]^{-1}, \tag{23}$$

where $A = (A_1^2 + A_2^2)$ and

$$\begin{cases} A_1 = 1 - \left(\dfrac{1}{\tau_R\omega}\right)^2 + 4\dfrac{\lambda c_v}{\tau_R}\left(q_{x_0}\dfrac{\partial \ln \lambda}{\partial \theta}\right)^{-2} \\ A_2 = \dfrac{2}{\tau_R\omega}\left[1 + 2\dfrac{\lambda c_v}{\tau_R}\left(q_{x_0}\dfrac{\partial \ln \lambda}{\partial \theta}\right)^{-2}\right], \end{cases} \tag{24}$$

for the model (5), whereas, for the model (3), from Eq. (22b) it follows

$$\begin{cases} A_1 = -\left(\dfrac{1}{\tau_R\omega}\right)^2 + 4\dfrac{\lambda c_v}{\tau_R}\left(q_{x_0}\dfrac{\partial \ln \lambda}{\partial \theta}\right)^{-2} \\ A_2 = \dfrac{4}{\tau_R\omega}\dfrac{\lambda c_v}{\tau_R}\left(q_{x_0}\dfrac{\partial \ln \lambda}{\partial \theta}\right)^{-2}. \end{cases} \tag{25}$$

Thus, once nonlinear terms are introduced in the transport equations, the speeds of thermal waves along a heat flux, arising from the models (3) and (5), will be always different, except for the case of low-frequency thermal-wave propagation.

III. HEAT PROPAGATION IN SYSTEMS WITH NONLINEAR GRADIENT-DEPENDENT TRANSPORT LAWS

In Sec. II we have started from linear evolution equations with the material functions dependent on θ and the nonlinearities arising from the temperature dependence of the material coefficients $c_v(\theta)$, $\lambda(\theta)$, and $\tau_R(\theta)$. Here, we show that nonlinear transport laws can be also obtained by introducing quadratic terms in the temperature gradient in the evolution equation for β [Eq. (2)]. These will introduce explicit nonlinear terms in the transport equation. For the sake of simplicity, in the present section, as well as in Sec. IV, the hypothesis of constant material functions will be made.

In Ref. 14, with an extension[25] of classical Liu[26] procedure and starting from the constitutive assumption in Eq. (1), it has been derived the following nonlocal evolution equation for the dynamical semiempirical temperature β:

$$\dot{\beta} = -\frac{1}{\tau_R}(\beta - \theta) - \frac{\chi}{\theta}\beta_{,i}\beta_{,i}. \tag{26}$$

Then, the combination of Eq. (26) with the constitutive assumption in Eq. (1) gives

$$\tau_R\dot{q}_i + q_i = -\lambda\theta_{,i} + \frac{2}{\theta}\frac{\tau_R}{c_v}q_k q_{k,i}, \tag{27}$$

once the third-order terms are neglected.[14]

Limiting ourselves to the one-dimensional case again (as before x means the longitudinal spatial coordinate), from Eqs. (6) and (27) we get the following hyperbolic equation for the nonequilibrium temperature θ:

$$\tau_R c_v\ddot{\theta} + c_v\dot{\theta} = \lambda\theta_{,xx} + \frac{2}{\theta}\tau_R q_{x_0}\dot{\theta}_{,x}, \tag{28}$$

where the quantity $2\tau_R q_{x_0}/\theta$ has been not neglected since it is taken as fixed parameter. Note that the relations above would no longer be true in the case of temperature-dependent material functions.

Supposing again the propagation of longitudinal plane temperature waves in Eq. (8), in the case of high-frequency perturbations from Eq. (28) one may derive the dispersion relation

$$k^2 - \frac{2}{\theta_0}\frac{\tau_R}{\lambda}q_{x_0}\omega k - \frac{\tau_R}{\chi}\omega^2 = 0, \tag{29}$$

whose form is very similar to that of Eq. (12a). Then, we obtain that the phase velocities $U^{\pm}$, in this case, result in

$$U^{\pm} = \sqrt{\frac{\chi}{\tau_R}\frac{1}{\sqrt{1+\Phi^2}\mp\Phi}}, \tag{30}$$

where $\Phi = [q_{x_0}/(c_v\theta_0)]\sqrt{\tau_R/\chi}$. The predictions in Eq. (30) point out that a small temperature pulse will travel with a different velocity in the direction of the heat flow (U^+) than in the opposite direction (U^-) also in this case. In particular, for $\Phi \ll 1$ Eq. (30) yields the following difference in the phase velocities:

$$U^- - U^+ = -\frac{2}{c_v\theta_0}q_{x_0}, \tag{31}$$

which becomes

$$U^- - U^+ = -\frac{5\pi^4}{6}\frac{M}{R\rho}\left(\frac{\theta_B}{\theta}\right)^3\frac{q_{x_0}}{\theta_0} \tag{32}$$

at low temperature, once the classical Debye expression for the specific heat per unit volume c_v has been used, namely, $c_v = (12/5\pi^4)(\theta/\theta_B)^3(R\rho/M)$, with θ_B as the Debye temperature, R as the gas constant, M as the molar mass, and ρ as the mass density. Note that, as mentioned in the previous section, in contrast with the prediction in Eq. (18), in this new case the phase velocity U^+ is greater than U^-. Up to first order in the steady-state heat flux, the modifications obtained in this section for the propagation speed [Eq. (31)] could be added to the modifications found in Sec. II for temperature-dependent material coefficients [Eq. (15)].

In the case of a nanowire made by silicon ($\bar{v} = 8.43\times10^3$ ms^{-1}, $\theta_B = 645$ K, $M_{\mathrm{Si}} = 28\times10^{-3}$ Kg mol^{-1}, $\rho_{\mathrm{Si}} = 2.33\times10^3$ Kg m^{-3}, and $R = 8.31$ JK^{-1} mol^{-1}) one may obtain the results in Table I. It is worth noticing that the difference in velocities in Eq. (31), which at very low temperature is high, decreases at higher temperature.

In the very general case, the dispersion relation given by Eq. (28) is

$$k^2 - 2\frac{\omega}{\lambda}\frac{\tau_R}{\theta_0}q_{x_0}k - \frac{\omega}{\chi}(\omega\tau_R - i) = 0, \tag{33}$$

which produces the following phase velocities:

$$U^{\pm} = \frac{\lambda}{\tau_R}\frac{\theta_0}{q_{x_0}}\left(\sqrt[4]{C}\sqrt{\frac{1}{2} + \frac{C_1}{2\sqrt{C}}} \pm 1\right)^{-1}, \tag{34}$$

being $C = C_1^2 + C_2^2$ with

TABLE I. Main results for the difference in the phase velocities $U^- - U^+$ in a silicon nanowire [Eq. (31)]. The value of the average heat flux q_{x_0} is different in the case of a nanowire at 30 K with respect to that of a nanowire at 300 K. However, the corresponding value of the nondimensional parameter Φ, which accounts for the approximation of Eq. (31), is the same in both cases. The relaxation time is $\tau_R = l/\bar{v}$. The values of the mean-free path l of phonons in silicon, for the different values of the temperature, have been obtained by experimental data.

θ_0 (K)	l (nm)	τ_R (s)	c_v ($\mathrm{Jm^{-3}\,K^{-1}}$)	λ ($\mathrm{Wm^{-1}\,K^{-1}}$)	q_{x_0} ($\mathrm{W\,m^{-2}}$)	Φ	$U^- - U^+$ ($\mathrm{m\,s^{-1}}$)
30	1.63×10^{4}	1.93×10^{-9}	1.71	7.85×10^{-2}	0.25×10^{2}	10^{-4}	-9.75×10^{-1}
300	0.4×10^{2}	4.74×10^{-12}	1.25×10^{6}	0.14×10^{3}	1.27×10^{3}	10^{-4}	-6.77×10^{-7}

$$\begin{cases} C_1 = 1 + \left(\dfrac{\theta_0}{q_{x_0}}\right)^2 \dfrac{\lambda c_v}{\tau_R}, \\ C_2 = \dfrac{1}{\omega\tau_R}\left(\dfrac{\theta_0}{q_{x_0}}\right)^2 \dfrac{\lambda c_v}{\tau_R}. \end{cases} \tag{35}$$

Finally, it is also interesting to explore how the boundary effects may contribute to the speed of heat waves. Indeed, in nanosystems the relative effects of area with respect to volume are relevant. In a nanowire, or in a thin layer, if it is not laterally isolated, the local balance of internal energy [Eq. (6)] is

$$\dot{e} = -q_{i,_i} - \frac{2\sigma}{r}(\theta - T_{\mathrm{env}}), \tag{36}$$

with the term in $(\theta - T_{\mathrm{env}})$ in its right-hand side representing the heat flux exchanged with the surrounding environment across the lateral walls of the system. It is in accordance with the Newton cooling law, being σ a suitable heat-exchange coefficient, r the radius, and T_{env} the temperature of the environment, which will be assumed to be constant. Thus, the previous evolution equation for θ [Eq. (28)] will be modified in

$$\tau_R c_v \ddot{\theta} + c_v\left(1 + 2\frac{\sigma}{r}\frac{\tau_R}{c_v}\right)\dot{\theta} = \lambda\theta_{,xx} + 2\frac{\tau_R}{\theta_0} q_{x_0}\dot{\theta}_{,x} - 2\frac{\sigma}{r}\left[\theta - T_{\mathrm{env}} + 2\tau_R\dot{\theta}\left(1 - \frac{T_{\mathrm{env}}}{\theta_0}\right)\right], \tag{37}$$

whose solution has been discussed by Jou *et al.*[27] in the linear regime, i.e., taking equal to zero the quadratic terms appearing in Eq. (26). It has been seen that for high frequencies, the phase velocity is not changed by the presence of the surface terms, and it is $U = U_0 = \sqrt{\chi/\tau_R}$. However, they modify the speed of thermal waves at relatively low frequencies. Here we will explore how the nonlinear term modifies the speeds in the presence of a heat flux. From Eq. (37) straightforward calculations point out that the phase velocities are still given by Eqs. (34), whereas one has

$$\begin{cases} C_1 = 1 + \left(\dfrac{\theta_0}{q_{x_0}}\right)^2 \left(\dfrac{\lambda c_v}{\tau_R} - 2\dfrac{\sigma}{r}\dfrac{\lambda}{\omega^2\tau_R^2}\right), \\ C_2 = \dfrac{\lambda}{\omega\tau_R}\left(\dfrac{\theta_0}{q_{x_0}}\right)^2 \left(\dfrac{c_v}{\tau_R} + 6\dfrac{\sigma}{r}\right), \end{cases} \tag{38}$$

instead of Eqs. (35).

In order to show the influence of the nonlinear terms, let us consider the high-frequency limit of Eq. (37). In this case, the phase velocities in Eq. (34) reduce to

$$U^{\pm} = \frac{\lambda}{\tau_R}\frac{\theta_0}{q_{x_0}}\left(\sqrt{1 + \frac{\lambda}{\tau_R}\left(\frac{\theta_0}{q_{x_0}}\right)^2} \pm 1\right)^{-1}, \tag{39}$$

which is different from that obtained by Jou *et al.*[27] Thus, the presence of the quadratic term in $\beta_{,i}$ in the evolution equation for the dynamical temperature β [Eq. (26)] gives a different phase velocity for q_{x_0} flowing in the same direction of the longitudinal plane temperature waves, Eq. (8), compared with q_{x_0} flowing in the opposite direction.

IV. NONLINEAR HYDRODYNAMIC ANALOGY

Phonon hydrodynamics is a regime of phonon heat transfer in which the role played by nonlocal effects becomes relevant as the size of a microdevice decreases[11] and the form of the nonlocal constitutive equation for the heat flux, expressed by the Guyer-Krumhansl equation[19,28,29]

$$\tau_R \dot{q}_i + q_i = -\lambda\theta_{,i} + l^2(q_{i,_{kk}} + 2q_{k,_{ki}}) \tag{40}$$

becomes very similar to the hydrodynamic equation for the velocity in a viscous fluid. In Eq. (40) l means a suitable coefficient accounting for the mean-free path of phonons. A comparison with kinetic theory of phonons shows that $l^2 = (9/5)\lambda\tau_N/c_v$, being τ_N the relaxation time of normal processes of interaction among the phonons. Macroscopic derivations of Eq. (40) have been obtained in weakly nonlocal nonequilibrium thermodynamics.[11,25,30–32] Indeed, if in Eq. (2) are introduced nonlocal effects as

$$\dot{\beta} = -\frac{1}{\tau_R}(\beta - \theta) + 3\frac{l^2}{\tau_R}\beta_{,ii}, \tag{41}$$

straightforward calculations allow to recover Eq. (40) in the case of constant material functions.

A generalization of the Guyer-Krumhansl equation (the basis for phonon hydrodynamics), incorporating nonlinear quadratic terms, analogous to the extra terms appearing in Eq. (27), corresponds to the following equation for β which generalizes Eq. (26) as

$$\dot{\beta} = -\frac{1}{\tau_R}(\beta - \theta) - \frac{\chi}{\theta}\beta_{,i}\beta_{,i} + \frac{3l^2}{\tau_R}\beta_{,ii}. \tag{42}$$

Coupled with Eq. (1) this yields the nonlinear Guyer-Krumhansl equation

$$\tau_R \dot{q}_i + q_i = -\lambda\theta_{,i} - \mu q_k q_{k,i} + l^2(q_{i,kk} + 2q_{k,ki}), \tag{43}$$

being $\mu = -2\tau_R/(c_v\theta)$.

Let us consider now a one-dimensional heat conductor and let x denote the position of the points of the system. Hence, the heat flux $\mathbf{q}$ is given by $\mathbf{q} = q_x(x)\mathbf{i}$, where q_x denotes the heat flux along the conductor and $\mathbf{i}$ is a unitary vector along the direction x. Then, in unsteady state and when q_x is negligible with respect to its spatial derivatives, Eq. (43) yields

$$\tau_R \dot{q}_x + \mu q_x q_{x,x} = -\lambda\theta_{,x} + 3l^2 q_{x,xx}. \tag{44}$$

The condition $q_x \ll l^2 q_{x,xx}$, necessary to neglect the heat flux q_x in Eq. (43), is satisfied, for instance, in one-dimensional nanowires where the characteristic length L is very small and $q_{x,xx} \approx q_x/L^2$. Equation (44) is analogous to the Navier-Stokes one, governing the evolution of a one-dimensional fluid, namely,

$$\rho(\dot{u}_x + u_x u_{x,x}) = -p_{,x} + \eta u_{x,xx}, \tag{45}$$

where ρ is the density of the fluid, u_x is its velocity along the longitudinal axis, η is the shear viscosity, and p is the pressure field. Such an analogy is evident in Grad's 14-moments theory,[33] where the first moment $\mathbf{p}$ is related to the heat flux by the constitutive equation $q_i = \bar{v}^2 p_i$.

The essential physics of a fluid is dictated by a competition between various phenomena, which is captured by a series of dimensionless numbers expressing their relative importance. Of all dimensionless numbers, the Reynolds number (Re) is the most important one. It relates the inertial forces to the viscous forces and it is given by

$$\mathrm{Re} = \frac{\rho u D}{\eta}, \tag{46}$$

where D means a typical length scale, as, for instance, the radius or diameter of a tube of the separation between two parallel walls. If Re is small the nonlinear term in Eq. (45) disappears whereas, as Re gets high this term cannot be neglected and it may destabilize the flow, resulting in unpredictable and irregular turbulent situation.

When these results are transferred to the heat-flow problem described by Eq. (44), along with Eq. (46), one may introduce the following thermal Reynolds number:

$$\mathrm{Re}_q = 2\frac{q_x \tau_R L}{\theta c_v l^2}. \tag{47}$$

That way, the stability of thermal flow can be analyzed by the methods of classical hydrodynamics.[34,35] In analogy with the hydrodynamic case, one may assume that a sudden decrease in the effective thermal conductivity arises for Re_q of the order 2300 (for cylindrical systems as nanowires), showing the nonlinear effects. From Eq. (47) it follows that the critical heat flux $q_{x,\mathrm{crt}}$ would be

$$q_{x,\mathrm{crt}} = 1150\,\theta c_v \bar{v}\left(\frac{l}{L}\right), \tag{48}$$

since the relaxation time τ_R is defined as $\tau_R = l/\bar{v}$. This equation points out that $q_{x,\mathrm{crt}}$ increases for high values of the ratio l/L, which is the well-known Knudsen number (Kn). Recall that Eq. (44) is valid for nanosystems, where the condition $L \ll l$ holds. In particular, for a silicon nanowire at 30 K and for Kn=10 the critical heat flux in Eq. (48) is $q_{x,\mathrm{crt}} = 4.98 \times 10^9$ W m^{-2}. It is worth to observe that for silicon at 30 K it holds $l_{\mathrm{Si}} = 1.63 \times 10^4$ nm, and Kn=10 would correspond to $L = 1.63 \times 10^3$nm. This value decreases very fast for lower temperature since it is easy to observe that $q_{x,\mathrm{crt}} \approx \theta^4$ because at low temperature, along with the Debye theory, $c_v \approx \theta^3$, as observed in Sec. III. That way, for $\theta = 3$ K and Kn=10 one would have $q_{x,\mathrm{crt}} = 4.98 \times 10^5$ W m^{-2}.

Equation (48) shows that nonlinear effects could become important at low temperature and moderate Knudsen numbers. We are not aware, for the moment, of the experimental results exhibiting these nonlinear effects but it may be understood that nanowires could provide a suitable testing ground for these effects.

V. CONCLUDING REMARKS

Heat waves are an interesting playground for modern transport theory because the behavior of high-frequency waves, or the incorporation of nonlinear effects, require nontrivial modifications in the transport equations, and of the entropy and temperature themselves, because they correspond to phenomena going beyond the local-equilibrium regime.

Here, we have compared two thermodynamic descriptions of heat relaxation going beyond the Fourier law. One of them is based on a dynamical temperature β and the other one on the Maxwell-Cattaneo equation. Note that the first description is in realm of rational thermodynamics, since the state space was enlarged by an internal variable together with its gradient while the heat flux has been assigned by a constitutive equation. The second description, instead, is in the framework of extended thermodynamics,[11,12] since the heat flux enters the state space and an evolution equation for it was given. Since, as shown in Ref. 27, the phase speed of thermal waves depends not only on the thermodynamic model, but on the material properties of the conductor too, in general it is not possible to decide what of the previous approaches leads to correct results. We feel that we should choose one of them depending on different situations. For constant material functions both descriptions yield identical results for thermal-wave propagation.[14] Thus, we have put our attention on nonlinear situations.

In Sec. II, we have assumed linear transport equations but with temperature-dependent coefficients. We have obtained the phase speed for thermal waves along a nonequilibrium steady state with a temperature gradient. Such wave speed turns out to be different in both descriptions, when the thermal conductivity depends on temperature. In the model based on a dynamical nonequilibrium temperature the high-frequency propagation speeds along the average heat flux and against it are different, whereas in the model based on the Maxwell-Cattaneo equation both speeds are equal.

In deriving Eqs. (7) and (10) we have neglected third-order terms. It seems important to investigate whether, neglecting specific terms on the level of the evolution equations, the compatibility of the considered models with second law of thermodynamics is still granted. We investigate this problem by considering the following two aspects: (1) if third-order terms are negligible in Eqs. (7) and (10), they should be also neglected in the balance of entropy; as a consequence of that, the form of the local-entropy production could change in such a way that second law is no longer respected. (2) Since Eqs. (7) and (10) contain the constitutive equations in a convolute way, the approximation of the evolution equations could result in an equivalent approximation of the constitutive relations which renders them incompatible with second law.

Let us start by investigating the problem (1). As far as the model with semiempirical temperature is concerned, which was developed within the frame of rational thermodynamics, we have exploited the Clausius-Duhem inequality

$$\sigma_s = -(\dot{\psi} + s\dot{\theta}) - \frac{1}{\theta} q_i \theta_{,i} \geq 0, \tag{49}$$

being $\psi = (e - \theta s)$ the Helmholtz free energy, by the generalized Coleman-Noll procedure developed in Ref. 36. We have obtained the following form for ψ:

$$\psi = \psi_0(\theta;\beta) + \frac{1}{2}\frac{\lambda \tau_R}{\theta \Gamma} \beta_{,i}\beta_{,i}, \tag{50}$$

with $\Gamma(\theta;\beta) = 1 + (\partial \ln \tau_R / \partial\theta)(\beta - \theta)$ while the entropy production reads

$$\sigma_s = \frac{\partial \psi}{\partial \beta}\frac{1}{\tau_R}(\beta - \theta) + \frac{\lambda}{\theta\Gamma}(\beta_{,i})^2, \tag{51}$$

(see Ref. 36 for more details). From Eqs. (1) and (50) we infer that the constitutive equations do not contain third-order terms. Moreover, by combining Eqs. (50) and (51), it is easy matter to show that, in the present case, the entropy production is not influenced by the approximation used in Eq. (7) since also σ_s does not contain third-order terms.

In the case of the model with Maxwell-Cattaneo equation, namely, that leading to Eq. (10), it can be recovered in the framework of extended thermodynamics,[11,12] by regarding the Maxwell-Cattaneo equation as a balance law for the heat flux with a particular form of the flux of heat flux.[25] In such a case, the state space is local and it is spanned by the variables $\{\theta;\mathbf{q}\}$. The exploitation of the entropy inequality

$$\sigma_s = \dot{s} + \left(\frac{q_i}{\theta}\right)_{,i} \geq 0, \tag{52}$$

leads to the conclusion that the constitutive equation for s is quadratic in $\mathbf{q}$ while the entropy production takes the form

$$\sigma_s = \frac{1}{\theta^2 \lambda} q_i q_i. \tag{53}$$

The right-hand side of Eq. (53) is nonnegative whatever the form of λ is and does not contain third-order terms. Hence, also in this case, σ_s is not influenced by the approximation used in Eq. (10).

Let us consider now the problem (2). In deriving Eq. (7) the sole constitutive equations to be considered are those for the internal energy and for the heat flux. The first one contributes only the first three terms appearing in the left-hand side of Eq. (7), which have been not neglected. On the other hand, the constitutive equation for the heat flux leads to the generalized Maxwell-Cattaneo Eq. (5), which has been calculated without any approximation. Moreover, Eq. (7) is obtained by calculating the time derivative of the balance of energy in Eq. (6) and the divergence of Eq. (5) and finally combining the obtained expressions. Doing that, one can see that the derivatives of Eqs. (5) and (6) contain third-order terms, which are not a consequence of the constitutive equations but of the chain rule. For the sake of simplicity, and only for that, we restricted ourselves to processes for which these terms are negligible. On the other hand, neglecting these terms, we did not assume different constitutive equations, because the same equations produce also first-order terms and second-order terms, which have been not neglected.

A similar analysis can be applied to the extended thermodynamic case. Here, the heat flux is not a constitutive quantity but enters the state space. The only constitutive equation to be considered in deriving Eq. (10) is that of the internal energy. As in the previous case, it contributes the first three terms in the left-hand side of Eq. (10). On the other hand, Eq. (10) has been obtained by combining the time derivative of the balance of energy and the divergence of the Maxwell-Cattaneo equation. Again, doing that, we have neglected third-order terms generated by the chain rule but conserved first- and second-order terms generated by the constitutive equation of the internal energy.

We conclude that, for both models considered in the present paper, third-order terms in the evolution equations can be neglected without compromising their compatibility with second law of thermodynamics. It is worth observing that, although such a result is true in our case, we cannot say that it is true in general.

Until now, we have analyzed the influence on the constitutive equations and on the local-entropy production of the approximation of the evolution equations up to second order. It is also important to investigate how the requirement of non-negative entropy production can influence the evolution equations. To this end, let us observe that it is not enough to decide the detailed form of the transport equations. For instance, in Eq. (53) the entropy production will be non-

TABLE II. Main results for the second-sound propagation. The propagation of longitudinal plane temperature waves [Eq. (8)] in the high-frequency limit has been considered. For the results below the identifications $2\Lambda=q_{x_0}(\sqrt{\tau_R/\lambda c_v})(\partial \ln \lambda/\partial\theta)$ and $\Phi=[q_{x_0}/(c_v\theta_0)]\sqrt{\tau_R/\chi}$ hold.

Constitutive assumptions	Evolution equation	Phase velocities
	Section II	
$q_i=-\lambda\beta_{,i};\lambda=\lambda(\theta);\ c_v=c_v(\theta);\ \tau_R=\tau_R(\theta);$	$\dot\beta=-(1/\tau_R)(\beta-\theta)$	$U^{\pm}=\sqrt{(\chi/\tau_R)}(1/\sqrt{1+\Lambda^2}\pm\Lambda)$
$\lambda=\lambda(\theta);\ c_v=c_v(\theta);\ \tau_R=\tau_R(\theta);$	$\tau_R\dot q_i+q_i=-\lambda\theta_{,i}$	$U^{+}=U^{-}=\sqrt{(\chi/\tau_R)}$
	Section III	
$q_i=-\lambda\beta_{,i};\lambda=\text{const};\ c_v=\text{const};\ \tau_R=\text{const};$	$\dot\beta=-(1/\tau_R)(\beta-\theta)-(\chi/\theta)\beta_{,i}\beta_{,i}$	$U^{\pm}=\sqrt{(\chi/\tau_R)}(1/\sqrt{1+\Phi^2}\mp\Phi)$

negative whatever is the temperature dependence of the thermal conductivity, provided this coefficient is positive; therefore, it cannot decide on the form of the temperature dependence. On the other hand, when higher-order terms in the fluxes or the gradients are incorporated to the transport equation, they yield corresponding higher-order terms in the entropy production. But these higher-order terms will be, in general, truncated approximations to a more complicated transport equation, and the positiveness of the corresponding truncated entropy production will only be able to state the limits of validity of the truncated expression, rather than the inconsistency of the nonlinear terms whose possible negative contribution to the entropy production could be compensated by a positive contribution of higher-order nonlinear terms.

Another way of measuring some difference between Eqs. (7) and (10) would be by using the techniques of thermal differential analysis. In these techniques, a sample of the material is homogeneously heated by means of an energy supply $r(t)$, in such a way that the local balance of specific internal energy [Eq. (6)] would be written as $\dot e=-q_{i,i}+r$. Since the term $r(t)$ is controlled and well known as a function of time, one measures the temperature $\theta(t)$ as a function of time. If the sample is small and thin enough it will have a homogeneous temperature, and Eqs. (7) and (10) will reduce to

$$\tau_R c_v\ddot\theta+c_v\dot\theta+\tau_R\left(\frac{\partial c_v}{\partial\theta}-\frac{c_v}{\lambda}\frac{\partial\lambda}{\partial\theta}\right)\dot\theta^2=r(t) \tag{54}$$

and

$$\tau_R c_v\ddot\theta+c_v\dot\theta+\tau_R\frac{\partial c_v}{\partial\theta}\dot\theta^2=r(t), \tag{55}$$

respectively. Another possibility of thermal differential analysis is to control $\theta(t)$, and imposing to the source term $r(t)$ the required time variation, to have the wanted time evolution $\theta(t)$. In particular, if one imposes that the temperature is increasing linearly with time ($\ddot\theta=0$), Eqs. (54) and (55), respectively, become

$$c_v\dot\theta+\tau_R\left(\frac{\partial c_v}{\partial\theta}-\frac{c_v}{\lambda}\frac{\partial\lambda}{\partial\theta}\right)\dot\theta^2=r(t) \tag{56}$$

and

$$c_v\dot\theta+\tau_R\frac{\partial c_v}{\partial\theta}\dot\theta^2=r(t). \tag{57}$$

By comparing with the values predicted by Eqs. (56) and (57) the required values of the source term $r(t)$ to have $\dot\theta=\text{const}$, the most suitable of these equations could be found. In Sec. III we have incorporated explicitly nonlinear terms, quadratic in the heat flux, in the transport equations, and have obtained their contribution to the phase speed. The speeds U^+ and U^- are still different from each other, and the difference could be added to the differences obtained in Sec. II. The main results obtained in Secs. II and III, concerning second-sound propagation in the high-frequency limit, are summarized in Table II.

In Sec. IV, we have compared a nonlinear Guyer-Krumhansl equation for phonon hydrodynamics with the nonlinear Navier-Stokes one and we have seen that it is possible to define a Reynolds number for the heat flux, analogous to that of fluid, which would describe a transition to a state analogous to turbulence with a sudden decrease in the effective thermal conductivity and increase of the fluctuations of the heat flux and we have made an estimation of this value in silicon nanowires.

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