On a Higher-Gradient Generalization of Fourier's Law of Heat Conduction

C. I. Christov

Dept. of Mathematics, University of Louisiana at Lafayette, Lafayette, LA 70504-1010, USA

Abstract. This paper deals with a possible generalization of Fourier's law that incorporates spatial memory into the constitutive relation. The integral and differential versions of the memory terms in the constitutive relation are discussed. It is shown that the asymptotically correct model contains the biharmonic operator as the vehicle for the higher-order heat diffusion that also accounts for the spatial memory of the processes. Different solutions in 1D, 2D and 3D are presented to show the applicability of the new model.

Keywords: Fourier Law, heat conduction, higher-gradient generalization.
PACS: 44.00.00, 44.10.+i, 51.20.+d, 44.05.+e

INTRODUCTION

The Fourier law of heat conduction [1] is one of the most important constitutive relations in continuum physics. In its simplicity it is a paragon of modeling in continuum physics. The main idea enshrined in Fourier's law is that the heat flux is proportional to the negative gradient of the scalar quantity under consideration: the temperature. If the scalar is the concentration, then it is called Fick's law. The Navier–Stokes model for viscous liquids contains a law of similar nature that establishes the relationship between the tensor of stresses (analog of the flux) and the rate of strain tensor, i.e., the symmetric part of the velocity gradient (analog of the temperature gradient). Linear-gradient constitutive laws are the basis for the majority of models in continuum mechanics, and they perform splendidly in engineering applications. However, despite its widespread usage, the model based on the Fourier law has several major shortcomings and has limited validity. Some of these shortcomings are discussed in this paper.

It is clear that a linear-gradient constitutive law can only be an approximation to a more realistic functional dependence between the physical quantities. And, despite the overall satisfactory performance, there exist situations where the Fourier-like models are not merely rough approximations but incorrect, in fact.

Fourier suggested a linear connection between the heat flux $\boldsymbol{q}$ and the temperature gradient ∇T, namely

$$\boldsymbol{q} = -\kappa \nabla T, \tag{1}$$

where $\kappa > 0$ is the coefficient of heat conduction, and the sign "$-$" is the thermodynamically-consistent choice that does not lead to self-concentration of heat but to dispersion of the latter. Introducing Eq. (1) into the balance law

$$\rho c_p \frac{\partial T}{\partial t} + \operatorname{div}\boldsymbol{q} = 0, \tag{2}$$

one gets the following equation for the temperature:

$$\frac{\partial T}{\partial t} = \lambda \Delta T, \qquad \lambda = \frac{\kappa}{\rho c_p}, \tag{3}$$

where ρ is the density, c_p is heat capacity, and λ is called the coefficient of thermal conductivity (see, e.g., [2, Pg.124]). The last equation is parabolic, and any disturbance introduced somewhere in the domain under consideration will propagate in space with infinite speed, i.e., it will be felt immediately everywhere in the domain. This is one of the well known shortcomings of the Fourier law, and appears to have first been noted by Maxwell [3, pg.260]. Such behavior, which is most apparent at low temperatures and/or in high heat-flux conditions [4, 5], and is believed to violate causality. The remedy for this paradox is known as the Maxwell–Cattaneo law for the heat flux, which is nonlocal in time and allows propagation of heat waves with finite speeds [6, 7, 4, 8]. The material invariance of the Maxwell–Cattaneo law is considered in [9].

Another set of paradoxes that are characteristic for all linear gradient constitutive laws is connected with the improper behavior of the solutions at infinity. In hydrodynamics, the 2D stationary solution with polar symmetry does not decay at infinity but rather diverges logarithmically. In heat conduction, this paradox did not attract enough attention in order to be given a name, but in fluid dynamics a similar setback is known as the Stokes paradox [10, 11]. The Stokes paradox is connected to the fact that *no solution* of the linear Stokes equations exists that satisfies both the boundary conditions at the rigid body (say, a cylinder) and the asymptotic boundary conditions. The better behaved potential in 3D (decaying as $1/r$ at infinity, rather than diverging as $\ln r$ in 2D) allows for the existence of a physically meaningful solution of the linearized equations, but then the asymptotic series for acknowledging the nonlinearity are divergent. This improper behavior of the solution is known as the paradox of Whitehead [11, §8.3]. In this case, the decay of the solution of the linearized problem is so slow that, when it is substituted into the neglected nonlinear terms, the latter dominate the retained linear terms at infinity.

All these setbacks are connected with the unrealistically fast diffusion from a boundary to infinity, and testify that the connection between the flux and the temperature gradient is more complex than the one given by the Fourier law.

In the present work, we set to outline the paradoxes connected with the linear Fourier law and propose a generalization of the latter that takes into account higher–order gradients. In doing so, we also address the issue of connection between Fourier's law and Newton's law of cooling.

PARADOXES CONNECTED WITH INFINITE DOMAINS

It is intuitively clear that if one has a body whose temperature is different (say, higher) from the surrounding medium, then the effect of the rapid change in the temperature should be felt appreciably only in a limited domain around the body. Quantitatively speaking, this domain can be small or large, but it should not extend to infinity and the temperature profile should decay relatively quickly when the spatial independent variable approaches infinity. A decay of order $1/r$ is too slow to explain the observed phenomena because the temperature at infinity should not be affected by the temperature of the body. At the same time, the heat flux measured through the body surface should have some specific finite value that is not equal to zero. Both these natural expectations are violated by the solutions to the classical heat equation, Eq. (3), because of the overindustrious transport mechanism embodied by the Fourier law.

To elucidate this point, we first consider the problem of heat conduction in a 1D vertical layer between the planes $x = 0$ and $x = L$. The left surface (the "body"), $x = 0$, has a temperature T_b while the right surface is kept at T_0. The b.v.p. reads

$$\lambda T_{xx} = 0, \quad T(0) = T_b, \quad T(L) = T_0.$$

The solution is

$$T(x) = T_b + \frac{T_0 - T_b}{L} x,$$

and the flux through the boundary $x = 0$ is given by

$$\lambda \left. \frac{dT}{dx} \right|_{x=0} = \lambda \frac{T_0 - T_b}{L}.$$

This expression asserts that if the gap between the two planar surfaces becomes infinite, then the heat flux through the left wall will vanish. This does not sound very convincing, from the practical point of view, even though it can be argued that after a very long time the whole space is heated up and the temperature assumes a linear profile with vanishing slope. However, this requires an infinite amount of energy to be radiated from the left boundary, and contradicts the empirically-established Newton law of cooling, which stipulates a non-vanishing heat flux from the surface. Some arguments can be made to justify the zero flux, e.g., in practice one never has an infinite surface, etc, but such arguments cannot be used in all cases. The model should be paradox-free.

A similar situation occurs in 2D and 3D, where the steady governing equation of the heat conduction reads

$$\lambda \frac{1}{r^\alpha} \frac{\partial}{\partial r} \left(r^\alpha \frac{\partial T}{\partial r} \right) = 0, \quad T(a) = T_b, \quad T(R) = T_\infty,$$

where $\alpha = 1$ in the 2D case, and $\alpha = 2$ in the 3D case.

For the 2D heat conduction, the time-independent solution of this b.v.p. and the flux through the body's surface are

$$T(r) = -\frac{T_b - T_\infty}{\ln R - \ln a} \ln r - \frac{T_\infty \ln a - T_b \ln R}{\ln R - \ln a}, \qquad \lambda \left. \frac{\partial T}{\partial r} \right|_{r=a} = \lambda \frac{T_\infty - T_b}{a(\ln R - \ln a)}.$$

Once again, we discover that if we remove the outer boundary to infinity, $R \to \infty$, then the flux from the cylinder vanishes, which contradicts experimental observations.

In 3D, the solution and the flux through the body's surface read:

$$T(r) = \frac{T_b - T_\infty}{R - a} \frac{aR}{r} + \frac{RT_\infty - aT_b}{R - a}, \qquad \lambda \left. \frac{\partial T}{\partial r} \right|_{r=a} = \lambda R \frac{T_\infty - T_b}{a(R - a)}.$$

Now, the limit $R \to \infty$ gives a non-vanishing heat flux from the sphere, and the solution decays at infinity. Similarly to Stokes' paradox, this decay is too slow, and, if one considers convection at very small Prandtl numbers, one finds that the slowly decaying solution makes the neglected terms larger than the retained terms. Moreover, like Stokes' paradox, the above heat-conduction paradox is much more severe in 2D than in 3D.

NEWTON'S LAW OF COOLING

Another clue pointing to the inadequacy of the Fourier law is furnished by so-called Newton's law of cooling. Originally, Newton's law of cooling was concerned with the cooling of a body with an average temperature, $\langle T \rangle$, due to heat exchange with the surrounding medium whose temperature is kept at different value, say, T_0. Then, the rate of cooling is stipulated to be proportional to the difference between the body's temperature and the ambient temperature, namely

$$\frac{\partial \langle T \rangle}{\partial t} = \chi (T_0 - \langle T \rangle), \tag{4}$$

where χ is the cooling coefficient, which is related to the heat exchange at the boundary of the body. The main idea in formulating this kind of approximate model is that the predominant change in the temperature of the surrounding medium takes place in a relatively thin layer adjacent to the body's surface. Then, T_0 has the meaning of an equilibrium temperature far from the body. As already mentioned, Newton's law of cooling was originally concerned with the averaged quantities, but it can be argued that it is, actually, a consequence of a similar law that holds for the local flux through a point of the surface, ∂D, of a body D, namely

$$\left.\frac{\partial T}{\partial n}\right|_{x \in \partial D} = \kappa_f (T_0 - T), \tag{5}$$

where T_0 is the temperature in the medium next to the surface and $\partial / \partial n$ stands for the normal to the surface directional derivative. The meaning of "next" is rather vague, and, in many formulations, T_0 is the temperature of the points well separated from the surface. In the sense of boundary layer, $T + 0$ is the temperature at the outer edge of the layer, which is, in fact, infinitely remote, in the asymptotic sense. Equation (5), which is related to the properties of the "outer" medium, is often considered as a boundary condition when solving Eq. (3).

Alongside Fourier's law, Newton's law of cooling is the other very well established empirical observation connected to heat transfer. For the medium outside of the body, whose temperature is being modeled, the coefficients in Newton's and Fourier's laws are independent of each other, which hints at the idea that the constitutive relationship between the heat flux and the temperature gradient may be more elaborate that the simple linear constitutive relation embodied by Fourier's law. In other words, we might be able to connect the coefficient in the right-hand side of Newton's law to the coefficients of the additional terms in the flux-gradient constitutive relation for the outer medium. It is clear that a self-consistent description of the processes requires that Newton's law of cooling, Eq. (5), be a limiting case of the solution of the heat conduction problem in the "outer" medium when the largest gradients of the temperature are in the layer adjacent to the "inner" surface.

HIGHER-GRADIENT GENERALIZATION OF FOURIER'S LAW

As with any higher-gradient generalization of a constitutive relation, in the present case one has two paths to follow: to add higher gradients of the temperature and/or to add gradients of the flux. In the former case, we will speak about "spatial retardation" of the temperature, while, in the latter case, we can use the coinage "spatial relaxation of the flux." This terminology comes from the analogy to viscoelastic liquids and the Maxwell–Cattaneo law. It is understood that the phenomenological model proposed here should be verified in the future by molecular-dynamics computations. In connection to this, we mention that fourth-order gradients of the temperature were argued in [12] as the manifestation of the effects of the so-called Knudsen layer in rarefied gases. In this paper, we limit ourselves to providing some general arguments for more complex constitutive relationships for the temperature, beyond the mere Fourier law.

When acknowledging both the retardation and relaxation (in the above introduced sense), and limiting oneself to gradients of the lowest two orders, one can generalize Eq. (1) as follows

$$\mathbb{Q} : \nabla\nabla \boldsymbol{q} + \mathbb{S} : \nabla \boldsymbol{q} + \boldsymbol{q} = -\kappa \nabla T + \mathbb{C} : \nabla\nabla T + \mathbb{K} : \nabla\nabla\nabla T, \tag{6}$$

where, in general, $\mathbb{C}, \mathbb{S}$ are third-rank tensors, and $\mathbb{K}, \mathbb{Q}$ are tensors of fourth rank. The colon sign ":" stands for the contraction of indices (without complicating further the notation, we presume as many contraction as necessary based on the rank of the tensors involved). Note that there is freedom to take a tensorial coefficient *in lieu* of κ in order to account for the possible anisotropy of the material, but this goes beyond the scope of the present work. Similarly, we stipulate that $\mathbb{S}$ is a tensor of third rank, while $\mathbb{Q}$ is a tensor of fourth rank. Using the standard expression for isotropic tensors of third and fourth rank, we can reduce Eq. (6) to the following

$$-\chi_2 \Delta \boldsymbol{q} + \boldsymbol{q} = -\kappa \nabla T + \kappa_2 \nabla \Delta T, \tag{7}$$

where χ_2 and κ_2 are some scalar coefficients called respectively the "spatial relaxation" and the "spatial retardation" coefficients. The signs are arbitrary at this point and are chosen as they are above for the sake of convenience. It is quite straightforward to show that if $\chi_2 = 0$, then the one must have $\kappa_2 \geq 0$ in order for the above model to be thermodynamically correct. Respectively, for $\kappa_2 = 0$ we must have $\chi_2 \geq 0$.

Here, we note that just like the Fourier law, given by Eq. (1), the constitutive relation given by Eq. (7) is once again an approximate expression. The general relation involves either an integral representation of the memory of the process (see Section) or an infinite number of higher-order derivatives. When resorting to a truncated version of the memory relationship, such as Eq. (1) or Eq. (7), one has to keep enough terms to ensure the correctness of the problem. In this instance, if $\kappa_2 < 0$, then one has to keep the sixth-order derivative of T and ensure that its coefficient has the proper sign. For example, in

[13] a spatially nonlocal model is proposed, but the second coefficient κ_2 has the wrong sign, from the thermodynamic point of view, but no higher-order terms are kept.

Finally, we note that the effect of the retardation term is such that, in the presence of a non-trivial gradient of the curvature of the temperature profile, the latter higher-order gradient acts as a resistance and reduces the evacuating effect of the gradient of the temperature itself. If the coefficients χ_2, κ_2 were not small, then the deviation from Fourier's law would be appreciable in all applications, which is not the case. Thus, one can safely conclude that both χ_2 and κ_2 are rather small quantities, and their effect is felt mostly in thin layers and/or in infinite domains.

THE GENERALIZED HEAT EQUATION AND ITS BOUNDARY CONDITIONS

After applying the Laplace operator to Eq. (3) and combining the result with Eq. (3), it can be shown that the generalized Fourier law, Eq. (7), yields the following equation for the temperature:

$$\frac{\partial T}{\partial t} - \chi_2 \frac{\partial \Delta T}{\partial t} = \lambda \Delta T - \lambda_2 \Delta\Delta T, \quad \lambda_2 = \frac{\kappa_2}{\rho c_p}. \tag{8}$$

Since, the coefficient of thermal conductivity λ has dimension $[\frac{\mathrm{m}^2}{\mathrm{s}}]$, the new coefficient λ_2 has dimension $[\frac{\mathrm{m}^4}{\mathrm{s}}]$, while χ_2 has dimension $[\mathrm{m}^2]$. As already mentioned, the new coefficients must both be very small (much smaller than λ) in order to leave the established results of heat transfer theory intact. While the term with coefficient χ_2 does not increase the spatial order of the equation, the term proportional to λ_2 does change the order. Even a very small λ_2 cannot be disregarded, especially in the boundary layers near surfaces, because it multiplies the highest spatial derivatives of the equation. In addition, the term with λ_2 may have cumulative effect that can change qualitatively the asymptotic behavior of the solution, even if the coefficient is small. See, e.g., [11] for discussion of perturbation methods.

Since Eq. (8) contains fourth derivatives, it requires an additional condition at every boundary. Thus, one can impose boundary conditions on both the function and its normal derivative, i.e., one can specify the temperature and enforce Newton's law of cooling simultaneously. If two conditions are not available, then, to find the ones that make the problem well-posed, we multiply Eq. (8) by $T(\boldsymbol{x},t)$ and integrate over the domain D in which the solution is sought. By means of integration by parts we arrive at

$$\begin{aligned}\frac{d}{dt}\iiint_D \left[T^2 + \chi_2(\nabla T)^2\right] \mathrm{d}^3\boldsymbol{x} = &-\iiint_D \left[\lambda(\nabla T)^2 + \lambda_2(\Delta T)^2\right] \mathrm{d}^3\boldsymbol{x} \\ &+ \oiint_{\partial D} \chi_2 T \frac{\partial}{\partial n}\frac{\partial T}{\partial t} \mathrm{d}\sigma + \oiint_{\partial D} T\Big(\frac{\partial T}{\partial n} - \lambda_2 \frac{\partial \Delta T}{\partial n}\Big)\mathrm{d}\sigma + \oiint_{\partial D} \frac{\partial T}{\partial n}\Delta T \mathrm{d}\sigma. \end{aligned} \tag{9}$$

Now, it is clear that the additional boundary conditions to be imposed in the absence of prescribed ones have to be dual to the missing conditions in the sense of canceling the surface integrals in Eq. (9). This means that if no condition is available for the temperature, then the natural boundary condition dual to the Dirichlet condition is

$$\left(\frac{\partial T}{\partial n} - \lambda_2 \frac{\partial \Delta T}{\partial n}\right)\Big|_{\boldsymbol{x}\in\partial D} = 0. \tag{10}$$

Respectively, when no boundary condition is available for the normal derivative of the temperature, one has to use the dual condition

$$\Delta T\Big|_{\boldsymbol{x}\in\partial D} = 0. \tag{11}$$

Thus, the following four combinations are possible (at different portions of the boundary, the combinations can be different):

$$T = f(\boldsymbol{x},t), \qquad \partial_n T = g(\boldsymbol{x},t), \tag{12}$$
$$T = f(\boldsymbol{x},t), \qquad \Delta T = 0, \tag{13}$$
$$\partial_n T = g(\boldsymbol{x},t), \qquad \partial_n T - \lambda_2 \partial_n \Delta T = 0, \tag{14}$$
$$\Delta T = 0, \qquad \partial_n T - \lambda_2 \partial_n \Delta T = 0, \tag{15}$$

for $\boldsymbol{x} \in \partial D$. Note that the last last two combinations would specify the solution only up to a constant.

ASYMPTOTICALLY EQUIVALENT FORMULATION

Now, we can make use of the fact that χ_2 and κ_2 are much smaller than unity and κ respectively. For the sake of convenience, we introduce the following notations: $\chi_2 = \bar{\chi}_2\varepsilon,\ \kappa_2 = \kappa\varepsilon$ and expand T and $\boldsymbol{q}$ in asymptotic series as

$$\begin{aligned} T(\boldsymbol{x},t) &= T_0(\boldsymbol{x},t) + \varepsilon T_1(\boldsymbol{x},t) + \varepsilon^2 T_2(\boldsymbol{x},t) + \ldots, \\ \boldsymbol{q}(\boldsymbol{x},t) &= \boldsymbol{q}_0(\boldsymbol{x},t) + \varepsilon \boldsymbol{q}_1(\boldsymbol{x},t) + \varepsilon^2 \boldsymbol{q}_2(\boldsymbol{x},t) + \ldots. \end{aligned} \tag{16}$$

Then, from Eq. (7), we get that

$$\boldsymbol{q}_0 = -\kappa \nabla T_0, \tag{17}$$
$$\boldsymbol{q}_1 = -\kappa \nabla T_1 + (1 + \kappa\bar{\chi}_2)\nabla\Delta T_0. \tag{18}$$

At this junction we have two options: to use Eq. (17) to express either $\boldsymbol{q}_0$ in terms of ∇T_0 or *vice versa*. The latter option will lead to losing the higher-order derivatives in the model, and the asymptotic reduction will lead to an equation that is of different type than the original Eq. (8). So, we use the former option and, after substituting $\boldsymbol{q}_0$ from Eq. (17) into Eq. (18), we arrive at the asymptotically equivalent constitutive law

$$\boldsymbol{q} = -\kappa \nabla T + \varepsilon(1 + \kappa\bar{\chi}_2)\nabla\Delta T + O(\varepsilon^2). \tag{19}$$

Thus, we have shown that in the asymptotic limit, $\varepsilon \ll 1$, we obtain a model with retardation only, but with a retardation coefficient that is increased by a quantity that is the product of the relaxation coefficient and the coefficient of heat conduction.

SOLUTIONS DEMONSTRATING THE APPLICABILITY

Let us begin with the 1D case. As already mentioned above, for classical Fourier's constitutive relation, there is no solution for the temperature that approaches (asymptotically) a given value at infinity while having a nontrivial flux at the body boundary. This paradox actually leaves no room for Newton's law of cooling. Now we investigate whether a solution of this type can exist for the generalized heat equation of the present work. Consider, for simplicity, a steady boundary value problem in a semi-infinite region $x \in [0, \infty)$ with the asymptotic b.c. at infinity and a Dirichlet b.c. at $x = 0$, namely

$$\begin{gathered} T'' - \varepsilon T'''' = 0, \\ T(0) = T_b, \quad T \to T_\infty \quad \text{for} \quad x \to \infty. \end{gathered} \tag{20}$$

At first sight, this boundary value problem appears to be underdetermined, but this may not be the case because an asymptotic b.c. is equivalent to an infinite number of conditions, so the problem may even be overdetermined in some cases.

The general solution of Eq. $(20)_1$ has the form

$$T(x) = c_1 e^{x/\sqrt{\varepsilon}} + c_2 e^{-x/\sqrt{\varepsilon}} + c_3 x + c_4.$$

The asymptotic b.c. are satisfied when $c_1 = c_3 = 0$ and $c_4 = T_\infty$. Only one constant is left unspecified, and the latter can be found by imposing the Dirichlet b.c. at $x = 0$. Thus, the solution of the b.v.p. given by Eq. (20) is

$$T(x) = T_b + (T_\infty - T_b)\left[1 - e^{-x/\sqrt{\varepsilon}}\right]. \tag{21}$$

This solution represents a temperature profile that makes good sense physically: it approaches T_∞ asymptotically and differs from that asymptotic value only in a thin region of order $\sqrt{\varepsilon}$ near the boundary with temperature $T_b \neq T_\infty$. And this is the situation that is imagined when Newton's law of cooling is argued (see, e.g., [14]). Actually, the above solution allows one to relate the constant in Newtion's law of cooling to the coefficient of the higher-gradient conductivity, namely

$$\left.\frac{\partial T}{\partial x}\right|_{x=0} = \frac{1}{\sqrt{\varepsilon}}(T_\infty - T_b).$$

In order to check whether the higher-gradient conductivity can resolve the paradox in 2D, we consider the boundary value problem

$$\begin{gathered} \frac{1}{r}\frac{d}{dr}\left\{r\frac{d}{dr}\left[T - \varepsilon\frac{1}{r}\frac{d}{dr}\left(r\frac{dT}{dr}\right)\right]\right\} = 0, \\ T(a) = T_b, \quad T \to T_\infty \quad \text{for} \quad r \to \infty, \end{gathered} \tag{22}$$

where $r = a$ is the cylindrical surface at which the emitting (Dirichlet) b.c. is imposed. The general solution to the above b.v.p. is

$$T(r) = c_1 K_0\left(\frac{r}{\sqrt{\varepsilon}}\right) + c_2 I_0\left(\frac{r}{\sqrt{\varepsilon}}\right) + c_3 \ln\left(\frac{r}{\sqrt{\varepsilon}}\right) + c_4, \tag{23}$$

where K_0 and I_0 are the modified Bessel functions of first and second kind. Once again, the asymptotic b.c. will be satisfied only when $c_2 = c_3 = 0$ and $c_4 = T_\infty$. The remaining integration constant is identified after satisfying the "emitting" b.c. at the cylinder's surface. Hence, the solution of Eq. (22) reads

$$T(r) = \frac{T_b - T_\infty}{K_0\left(a/\sqrt{\varepsilon}\right)} K_0\left(r/\sqrt{\varepsilon}\right) + T_\infty. \tag{24}$$

Here, it is important to note, that the asymptotic behavior for $r \to \infty$ of the solution given by Eq. (24) is $r^{-0.5}e^{-r/\sqrt{\varepsilon}}$, i.e., it decays slightly faster than exponentially, which is in full agreement with the decay rate in the 1D case. Recall that, under the classical theory, both the 1D and 2D cases exhibited paradoxical behavior.

Then, at the rigid boundary the normal gradient is given by

$$\left.\frac{\partial T}{\partial r}\right|_{r=b} = (T_\infty - T_b)\frac{K_1\left(a\sqrt{\varepsilon}\right)}{\sqrt{\varepsilon}K_0\left(a\sqrt{\varepsilon}\right)} = \frac{1}{\sqrt{\varepsilon}}(T_\infty - T_b)\left[1 + O(\sqrt{\varepsilon})\right]. \tag{25}$$

Here, we have made use of the fact that, for $\sqrt{\varepsilon} \ll 1$ and $a \sim 1$, the arguments of the Bessel functions are pretty large and the ratio can be approximately replaced by the limit which is equal to unity. Thus, we get the same flux through the boundary and the same Newton law as in the 1D case.

Without going into details, we mention that the solution of the above b.v.p. problem in 3D reads (ρ is the spherical coordinate)

$$T(\rho) = (T_b - T_\infty)\frac{a}{\rho}e^{-(\rho-a)/\sqrt{\varepsilon}} + T_\infty, \qquad \left.\frac{\partial T}{\partial \rho}\right|_{\rho=a} = \frac{1}{\sqrt{\varepsilon}}(T_\infty - T_b)\left(1 + \sqrt{\varepsilon}\right). \tag{26}$$

Here we have discarded a solution of type $1/r$ because of its slow decay at infinity.

THE GENERAL NONLOCAL RELATIONSHIPS

Adding higher-order derivatives in a constitutive relation amounts to acknowledging some memory of the process. This is rather obvious in the Maxwell–Cattaneo law. Since, in this paper, we consider only spatial higher-order derivatives we use the coinage "spatial memory." A nonlocal constitutive relationship with retardation memory assumes that to the flux, $\boldsymbol{q}$, at a certain point in space contributes to the temperature gradients, $\boldsymbol{G} \equiv \nabla T$, in the vicinity of this point (with different weights, of course). Respectively, to the temperature gradient at a given point, the fluxes from all adjacent points contribute with certain weights. In 1D, this statement can be expressed formally as

$$-\kappa \int_{-\infty}^{\infty} M(\zeta)G(x-\zeta)\mathrm{d}\zeta = \int_{-\infty}^{\infty} K(\xi)q(x-\xi)\mathrm{d}\xi. \tag{27}$$

Note that, in 1D, the temperature gradient, G, and flux, q, are scalar quantities. The (local) Fourier law is recovered from Eq. (27) when the kernels are Dirac delta functions, namely $M(\zeta) = \delta(\zeta)$ and $K(\xi) = \delta(\xi)$.

It is clear that the kernels K and M must have certain basic properties. First, they have to be positive because otherwise the model can lead to the increase in time of the L^2 norm of the quantity called temperature. In other words, the existence of negative portions of the kernels can violate the second law of thermodynamics. Second, the kernels must be even functions, since the temperature profile and the flux to the left or to the right of the point x must have the same "right" to influence the heat flux and temperature gradient at x, respectively. Third, $K(\xi)$ and $M(\zeta)$ must decay fast enough as $\xi,\zeta \to \infty$. At the same time, we have the normalization condition $\int_{-\infty}^{\infty} K(\xi)\mathrm{d}\xi = \int_{-\infty}^{\infty} M(\zeta)\mathrm{d}\zeta = 1$. A typical function having these properties is the exponential one. Thus, we stipulate

$$K(\xi) = \beta_q e^{-\beta_q^{-1}|\xi|}, \quad M(\zeta) = \beta_T e^{-\beta_T^{-1}|\zeta|}, \quad \beta_q, \beta_T > 0. \tag{28}$$

The above functions become Dirac delta functions in the limit of vanishingly short memory, i.e., as $\beta_q, \beta_T \to 0$.

Next, expanding $q(x-\xi)$ into Taylor series, we get

$$\begin{aligned}
&\int_{-\infty}^{\infty} K(\xi)\Big[q(x) - \xi q'(x) + \frac{\xi^2}{2}q''(x) - \frac{\xi^3}{6}q'''(x) + \frac{\xi^4}{24}q^{(4)}(x) + \cdots\Big]\mathrm{d}\xi \\
&= \Big[\int_{-\infty}^{\infty} K(\xi)\mathrm{d}\xi\Big]q(x) + \Big[\int_{-\infty}^{\infty} \frac{\xi^2}{2}K(\xi)\mathrm{d}\xi\Big]q''(x) + \Big[\int_{-\infty}^{\infty} \frac{\xi^4}{24}K(\xi)\mathrm{d}\xi\Big]q^{(4)}(x) + \cdots \\
&= q + \nu_2 q'' + \nu_4 q^{(4)} + \cdots, \qquad \text{where} \quad \nu_{2n} \overset{\text{def}}{=} \int_{-\infty}^{\infty} \frac{\xi^{2n}}{(2n)!}K(\xi)\mathrm{d}\xi.
\end{aligned} \tag{29}$$

We call these coefficients the "relaxation lengths" in an analogy to the "relaxation time" present in the Maxwell–Cattaneo law. For this particular choice of memory function, given in Eq. (28), we get the following asymptotic orders

$$\nu_{2n} = \varepsilon_q^n, \quad \varepsilon = \beta^2. \tag{30}$$

Note that, for memory functions that are more complex than the above assumed exponential type, the relationship between ν_{2n} and ε^n can involve coefficients different from unity. We set, for definiteness, $\nu_{2n} = \bar{\nu}_{2n}\varepsilon^n$, where $\bar{\nu}_{2n}$ is a coefficient of order of unity. A similar derivation for μ_{2n} yields

$$\begin{aligned}
&-\kappa\int_{-\infty}^{\infty} M(\zeta)\Big[G(x) - \zeta G'(x) + \frac{\zeta^2}{2}G''(x) - \frac{\zeta^3}{6}G'''(x) + \frac{\zeta^4}{24}G^{(4)}(x) + \cdots\Big]\mathrm{d}\zeta \\
&\qquad = -\kappa[G + \mu_2 G'' + \mu_4 G^{(4)} + \cdots] = -\kappa[T' + \mu_2 T''' + \mu_4 T^{(5)} + \cdots],
\end{aligned} \tag{31}$$

where

$$\mu_{2n} \overset{\text{def}}{=} \int_{-\infty}^{\infty} \frac{\zeta^{2n}}{(2n)!}M(\zeta)\mathrm{d}\zeta \tag{32}$$

will term the "retardation lengths."

Now, we can rewrite Eq. (27) as follows

$$-\kappa[T' + \mu_2 T''' + \mu_4 T^{(5)} + \cdots] = q + \nu_2 q'' + \nu_4 q^{(4)} + \cdots. \tag{33}$$

It is very important to outline the applicability of the above differential law in the framework of the asymptotic approximation when higher-order terms are neglected because of their smallness. First, we observe that *all* retardation and relaxation coefficients are positive because the kernels are positive and so are the terms ξ^{2n} and ζ^{2n}. Thus, one cannot consider only retardation or only relaxation memory in the differential approximation, even though the general constitutive relation with memory, Eq. (27), leads to a well-posed problem even if only one of the kernels is a Dirac delta function. If we start from Eq. (33) and disregard the relaxation lengths $\nu_2, \nu_4, \ldots$, then we cannot truncate the right-hand side at the term multiplied by μ_2, despite of the fact that $\mu_4, \ldots$ are small. While the integral formulation with positive kernels always gives a mathematically correct model, the truncated differential approximations are never correct unless an infinite number of higher-order derivatives is retained. If one does neglect these coefficients, then one will arrive at Eq. (8) with $\chi_2 = 0$ and $\lambda_2 < 0$, and this case has already been identified as incorrect. Alternatively, if one neglects all retardation and relaxation coefficients for $n \geq 2$, then one will have Eq. (8) with $\chi_2 < 0$ and $\lambda_2 = 0$, which is once again an incorrect model (in the sense of Hadamard).

This argument shows that there are limitations on the applicability of the differential constitutive relation given by Eq. (33). The first one is that the respective retardation and relaxation coefficients have the be of the same order and one should always keep the same number of them on each side of Eq. (33). This, however, is not enough because if we truncate the latter at ν_2 and μ_2 we arrive at a higher–order diffusion equation that can be correct only if the magnitudes of μ_2 and ν_2, are related appropriately. Yet, we show, here, that a correct differential approximation *can* be obtained.

In order to elucidate how one finds the relationship between the values of the retardation and relaxation coefficients, we take advantage of the fact that these coefficients must be very small, and we consider the following asymptotic expansions:

$$q = q_0 + \varepsilon q_1 + \varepsilon^2 q_2 + \cdots, \qquad T = T_0 + \varepsilon T_1 + \varepsilon^2 T_2 + \cdots. \tag{34}$$

Setting

$$\nu_2 = \varepsilon, \quad \mu_2 = d\varepsilon,$$

and introducing Eq. (34) in Eq. (33), we get for the first two terms:

$$q_0 = -\kappa T_0', \quad q_1 = -q_0'' - d\kappa T_0''' - \kappa T_1'. \tag{35}$$

Introducing Eq. $(35)_1$ into Eq. $(35)_2$ we find that

$$q_1 = -\kappa T_1' + \kappa(1-d)T_0''',$$

which gives

$$q_0 + \varepsilon q_1 = -\kappa(T_0' + \varepsilon T_1') + \varepsilon\kappa(1-d)(T_0''' + \varepsilon T_1''') + O(\varepsilon^2).$$

Finally, within the selected asymptotic order, we have the following constitutive relation

$$q = -\kappa T' + \kappa\hat{\varepsilon}T''', \qquad \hat{\varepsilon} = \varepsilon(1-d). \tag{36}$$

Thus, we have shown that the full constitutive relation is asymptotically equivalent to a law with spatial retardation. Introducing Eq. (36) into Eq. (2) gives Eq. (8) with $\chi_2 = 0$

which is thermodynamically consistent only if $d < 1$, i.e., when $\hat{\varepsilon} > 0$. It is clear that when $d > 1$, one can reverse the above asymptotic derivation and to obtain, once again, Eq. (8), this time with $\lambda_2 = 0$ and $\chi_2 > 0$, which is correct in the sense of Hadamard. Therefore, we have found the way to derive a thermodynamically consistent higher-gradient model for an arbitrary relation between relaxation and retardation lengths. If for a particular material $d < 1$, the correct higher-order model involves a biharmonic term, while for $d > 1$ the higher-order term is the time derivative of the Laplacian.

CONCLUSIONS

Some shortcomings of the classical models of heat conduction based on Fourier's law are discussed and the argument is put forward that they can be alleviated if a more general constitutive relation is used, in which higher-order gradients of heat flux and temperature are allowed. The higher–order gradients of the flux are called the "spatial relaxation," while the higher-order gradients of the temperature are termed the "spatial retardation." The higher-gradient relations are also considered from the point of view of constitutive relations for processes with spatial memory. This leads to a parabolic equation, containing biharmonic diffusion operators, and the conditions for the correctness of the respective initial-boundary value problems are established. The relation of the higher-gradient terms to the so-called Newton law of cooling is also discussed. Examples of the application of the new model to heat conduction in unbounded domains are presented.

ACKNOWLEDGMENTS

The author is indebted to Ivan Christov for carefully reading the manuscript and making a number of valuable suggestions.

REFERENCES

1. H. S. Carslaw and J. C. Jaeger, *Conduction of Heat in Solids*, Oxford University Press, New York, 1959, 2nd ed.
2. L. C. Thomas, *Fundamentals of Heat Transfer*, Prentice-Hall, Englewood Cliffs, New Jersey, 1980.
3. J. C. Maxwell, *Theory of Heat*, Longmans, Green, and Co., London, 1885.
4. D. S. Chandrasekharaiah, *Appl. Mech. Rev.* **39**, 355–376 (1986).
5. W. Dreyer and H. Struchtrup, *Cont. Mech. Thermodyn.* **5**, 3–50 (1993).
6. D. D. Joseph and L. Preziosi, *Rev. Mod. Phys.* **61**, 41–73 (1989).
7. D. D. Joseph and L. Preziosi, *Rev. Mod. Phys.* **62**, 375–391 (1990).
8. D. Jou, J. Casas-Vázquez, and G. Lebon, *Rep. Prog. Phys.* **62**, 1035–1142 (1998).
9. C. I. Christov and P. M. Jordan, *Phys. Rev. Lett.* **94**, 154301 (2005).
10. G. Birkhoff, *Hydrodynamics. A Study in Logic, Fact and Similitude*, Princeton University Press, New Jersey, 1960.
11. M. Van Dyke, *Perturbation Methods in Fluid Mechanics*, Academic Press, New York, 1964.
12. L. S. Pan, D. Xu, J. Lou, and Q. Yao, *Europhys. Lett.* **73**, 846–850 (2006).
13. S. I. Sobolev, *Phys. Rev. E* **50**, 3255–5588 (1994).
14. S. Whitaker, *Fundamental Principles of Heat Transfer*, Pergamon Press, New York, 1977.