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Macroscopic heat transport equations and heat waves in nonequilibrium states

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HIGHLIGHTS

- A generalized heat transport equation including nonlinear, nonlocal and relaxation terms is proposed.
- Heat wave propagations are investigated systematically in nonequilibrium steady states.
- The phase (or front) speed of heat waves is intimately related to the nonlinear and nonlocal terms.

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ABSTRACT

Heat transport may behave as wave propagation when the time scale of processes decreases to be comparable to or smaller than the relaxation time of heat carriers. In this work, a generalized heat transport equation including nonlinear, nonlocal and relaxation terms is proposed, which sums up the Cattaneo–Vernotte, dual-phase-lag and phonon hydrodynamic models as special cases. In the frame of this equation, the heat wave propagations are investigated systematically in nonequilibrium steady states, which were usually studied around equilibrium states. The phase (or front) speed of heat waves is obtained through a perturbation solution to the heat differential equation, and found to be intimately related to the nonlinear and nonlocal terms. Thus, potential heat wave experiments in nonequilibrium states are devised to measure the coefficients in the generalized equation, which may throw light on understanding the physical mechanisms and macroscopic modeling of nanoscale heat transport.

1. Introduction

Heat waves contribute to heat transport in fast processes besides the usual diffusive transport described by Fourier's law, and on the other hand, they may provide new experimental tools for the analysis of physical systems [1–10]. Recent investigations of heat transport in carbon nanotubes [11–13] and in graphene sheets or nano-ribbons [14–16] have declared the role of several non-Fourier features, related to a combined heat transfer in diffusive form and in form of heat waves. For instance, in Ref. [14] the authors studied the effects of a rapid cooling of four layers of carbon atoms at one end of a graphene nano-ribbon, which leads to rapid propagation of thermal perturbation, especially at the early period. They observed temperature responses described by the following generalized heat transport equation:

$$\frac{1}{\alpha}\frac{\partial T}{\partial t}+\frac{\tau_q}{\alpha}\frac{\partial^2 T}{\partial t^2}=\nabla^2 T+\tau_\theta\frac{\partial}{\partial t}\left(\nabla^2 T\right), \quad (1)$$

with α the thermal diffusivity, and τ_q and τ_θ two phase lags. In particular, they found $\tau_q = 1.85$ ps, $\tau_\theta = 1.01$ ps and $\alpha = 1.44 \times 10^{-5}$ m^2/s for a ribbon of length 14.9 nm. Although these values are very small, they are measurable by current experimental techniques. This work is mentioned as an example of generalized heat transport equations not only beyond Fourier's law ($\tau_q = \tau_\theta = 0$) but also beyond Cattaneo–Vernotte (C–V) law [17,18] ($\tau_\theta = 0$), one of the well-known equations in the description of heat waves [1,7]. The need to go beyond C–V law in the analysis of actual fast thermal processes motivates the current interest in exploring generalized heat transport equations.

In the present work, a generalized heat transport equation is proposed, which incorporates nonlinear and nonlocal terms into

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the C–V law. It sums up many previous macroscopic models for nanoscale heat transport [9] as special cases, allowing a comparison between their respective physical consequences in the propagation of heat waves. Usually, heat wave propagation was studied around equilibrium states [11–16,19–22]. Propagation of heat waves in nonequilibrium steady states has been only considered for some particular cases [23–28]. However, the nonlinear terms often neglected in generalized heat transport models have an intimate relevance to wave propagation in nonequilibrium steady states [26–28]. Thus the present work generalizes much the few previous works on this issue, and gives rise to some new features of heat waves along nonequilibrium steady states based on the proposed generalized equation. In addition, nanotechnology opens new perspectives to this problem, because it becomes possible to study the speed of thermal perturbations along carbon nanotube or graphene ribbons with their ends kept at different temperatures, thus imposing a controlled non-vanishing average heat flux along them.

The remainder of this article is organized below. In Section 2, the generalized heat transport equation is introduced, with a summary of how one may recover from it diverse heat transport equations of existing macroscopic models. Besides, the kinetic theory and thermodynamic foundations are also discussed for the generalized heat transport equation. In Section 3, the influences of nonlinear and nonlocal terms in the generalized equation are systematically studied on the phase speed of heat waves or front speed of heat pulse perturbations around nonequilibrium steady state. In Sections 4 and 5, discussions and concluding remarks are made.

2. A generalized heat transport equation

A generalized heat transport equation including nonlinear and nonlocal terms as well as a relaxation term is proposed as:

$$\tau\frac{\partial \mathbf{q}}{\partial t}+\mathbf{q}=-\lambda\nabla T+m_1\mathbf{q}\nabla\cdot\mathbf{q}+m_2\mathbf{q}\cdot\nabla\mathbf{q}+m_3\nabla\mathbf{q}^2+m_4\nabla^2\mathbf{q}+m_5\nabla\left(\nabla\cdot\mathbf{q}\right)+m_6\mathbf{q}\left(\mathbf{q}\cdot\nabla T\right)+m_7\mathbf{q}^2\nabla T, \tag{2}$$

where τ is the relaxation time of heat flux, λ is the thermal conductivity, and $m_i(T)$ ($i = 1, 2, \ldots, 7$) are temperature dependent coefficients to be identified below. In physical views, the main motivation in incorporating these terms originates in the analysis of nanosystems, where the spatial gradients of physical quantities such as temperature and heat flux may be extremely large due to the minute size of the system. On the other hand, the temporal derivative of heat flux may be extremely high in the fast local heating of a sample by intense and narrow laser beams. Eq. (2) contains particular cases of many previous macroscopic models for nanoscale heat transport, and provides a common ground for a comparison between them.

To recover the classical Fourier's law, all the terms in τ and m_i are vanishing whereas for the C–V law only the relaxation term is kept. The coefficients m_i are identified through comparing Eq. (2) to the heat transport equations respectively in dual-phase-lag (DPL) model [10]:

$$\tau_q\frac{\partial \mathbf{q}}{\partial t}+\mathbf{q}=-\lambda\nabla T-\lambda\tau_T\frac{\partial}{\partial t}\left(\nabla T\right), \tag{3}$$

with τ_q, τ_T the phase lags of heat flux and temperature gradient, in Guyer–Krumhansl (G–K) model [29] (phonon hydrodynamics model [30]):

$$\tau_{\mathrm{R}}\frac{\partial \mathbf{q}}{\partial t}+\mathbf{q}=-\lambda\nabla T+l^2\left[\nabla^2\mathbf{q}+2\nabla(\nabla\cdot\mathbf{q})\right], \tag{4}$$

with $l^2=\tau_{\mathrm{N}}\tau_{\mathrm{R}}v_{\mathrm{g}}^2/5$, τ_{N}, τ_{R} the relaxation times of phonon normal (N) and resistive (R) processes and v_{g} the average phonon group speed, in the nonlinear G–K model [28]:

$$\tau_{\mathrm{R}}\frac{\partial \mathbf{q}}{\partial t}+\mathbf{q}=-\lambda\nabla T+\frac{2}{T}\frac{\tau_{\mathrm{R}}}{C_V}\mathbf{q}\cdot\nabla\mathbf{q}+l^2\left[\nabla^2\mathbf{q}+2\nabla(\nabla\cdot\mathbf{q})\right], \tag{5}$$

with C_V the heat capacity per unit volume, and in the thermon gas model [31,32]:

$$\tau_{\mathrm{T}}\frac{\partial \mathbf{q}}{\partial t}+\mathbf{q}=-\lambda\nabla T-\frac{\tau_{\mathrm{T}}}{C_VT}\mathbf{q}\nabla\cdot\mathbf{q}-\frac{\tau_{\mathrm{T}}}{C_VT}\mathbf{q}\cdot\nabla\mathbf{q}+\frac{\tau_{\mathrm{T}}}{C_VT^2}\mathbf{q}\left(\mathbf{q}\cdot\nabla T\right), \tag{6}$$

with τ_{T} the relaxation time of thermon gas. Note that to recover Eq. (3) in the DPL model, the energy balance equation is supplemented [6]:

$$C_V\frac{\partial T}{\partial t}=-\nabla\cdot\mathbf{q}, \tag{7}$$

and the mixed partial derivative of temperature in Eq. (3) is reformulated as:

$$\frac{\partial}{\partial t}\left(\nabla T\right)=\nabla\left(\frac{\partial T}{\partial t}\right)=-\frac{1}{C_V}\nabla\left(\nabla\cdot\mathbf{q}\right). \tag{8}$$

The relations between the coefficients in Eq. (2) and those in the previous heat transport models are thoroughly summarized in Table 1. Besides, the terms in Eq. (2) with coefficients m_3 and m_7 not explicitly correlated to previous models could be got from a nonequilibrium temperature θ dependent on heat flux. The nonequilibrium temperature is obtained in extended irreversible thermodynamics as $\theta^{-1}\equiv\partial s/\partial u$ with $s\equiv s\left(u,\mathbf{q}\right)$ a generalized entropy dependent on u and $\mathbf{q}$, and becomes [6]:

$$\theta^{-1}=T^{-1}-\frac{1}{2}\frac{\partial}{\partial u}\left(\frac{\tau}{\rho\lambda T^2}\right)\mathbf{q}\cdot\mathbf{q}. \tag{9}$$

Substitution of Eq. (9) into an extended Fourier's law $\mathbf{q}=-\lambda\nabla\theta$ with an approximation $\theta\approx T+\xi\left(T\right)\mathbf{q}^2$ ($\xi\left(T\right)\equiv\frac{1}{2}T^2\partial\left(\tau/\rho\lambda T^2\right)/\partial u$ for brevity) gives rise to:

$$\mathbf{q}=-\lambda\left(1+\frac{\partial\xi}{\partial T}\mathbf{q}^2\right)\nabla T-\lambda\xi\nabla\mathbf{q}^2. \tag{10}$$

Thus the coefficients are identified as: $m_7=-\lambda\partial\xi/\partial T$, and $m_3=-\lambda\xi$. The terms in m_3 and m_7 could be logically incorporated as additional terms into the nonlinear G–K model equation (5) through the temperature gradient term, but usually they are not considered for simplicity because of their negligible effect.

Therefore the generalized heat transport equation (2) contains in a compact way the heat transport equations of diverse previous macroscopic models. Furthermore, these terms with coefficients m_i in Eq. (2) are not merely written in a phenomenological way, but actually deeply rooted in the kinetic theory of phonons [33,34]. The following heat transport equation has been derived from phonon Boltzmann equation by either maximum entropy [35] or Grad's type [36] moment methods and Chapman–Enskog expansion within zeroth-order approximation [9,37]:

$$\tau_{\mathrm{R}}\frac{\partial \mathbf{q}}{\partial t}+\mathbf{q}=-\lambda\nabla T-\frac{3\tau_{\mathrm{R}}}{C_V}\nabla\cdot\frac{\langle\mathbf{q}\mathbf{q}\rangle}{2T\left[1+\sqrt{1-\frac{3}{4}\left(q/v_{\mathrm{g}}C_VT\right)^2}\right]}, \tag{11}$$

where the deviatoric part of the tensor $\mathbf{q}\mathbf{q}$ denotes $\langle\mathbf{q}\mathbf{q}\rangle=\mathbf{q}\mathbf{q}-\frac{1}{3}\mathbf{q}^2\mathbf{I}$, with $\mathbf{I}$ the unit tensor. For relatively small heat flux ($q/v_{\mathrm{g}}C_VT\ll 1$), Eq. (11) is approximated as:

$$\tau_{\mathrm{R}}\frac{\partial \mathbf{q}}{\partial t}+\mathbf{q}=-\lambda\nabla T-\frac{3\tau_{\mathrm{R}}}{4C_V}\nabla\cdot\frac{\langle\mathbf{q}\mathbf{q}\rangle}{T}. \tag{12}$$

Table 1
Relations between the coefficients in the generalized heat transport equation (2) and in previous macroscopic models for nanoscale heat transport.

	Fourier's law	C–V law	DPL model equation (3)	G–K model equation (4)	Nonlinear G–K model equations (5), (10)	Thermon gas model equation (6)
τ	0	τ	τ_q	τ_R	τ_R	τ_T
m_1	0	0	0	0	0	$-\frac{\tau_T}{C_V T}$
m_2	0	0	0	0	$\frac{2}{T}\frac{\tau_R}{C_V}$	$-\frac{\tau_T}{C_V T}$
m_3	0	0	0	0	$-\lambda\xi$	0
m_4	0	0	0	l^2	l^2	0
m_5	0	0	$\alpha\tau_T$	$2l^2$	$2l^2$	0
m_6	0	0	0	0	0	$\frac{\tau_T}{C_V T^2}$
m_7	0	0	0	0	$-\lambda\partial\xi/\partial T$	0

With a full expansion of the second term on the rightside, Eq. (12) is reformulated as:

$$\tau_R \frac{\partial \mathbf{q}}{\partial t} + \mathbf{q} = -\lambda\nabla T - \frac{3}{4}\frac{\tau_R}{C_V T}\mathbf{q}\nabla\cdot\mathbf{q} - \frac{3}{4}\frac{\tau_R}{C_V T}\mathbf{q}\cdot\nabla\mathbf{q} + \frac{1}{4}\frac{\tau_R}{C_V T}\nabla\mathbf{q}^2 + \frac{3}{4}\frac{\tau_R}{C_V T^2}\mathbf{q}\left(\mathbf{q}\cdot\nabla T\right) - \frac{\tau_R}{4C_V T^2}\mathbf{q}^2\nabla T. \quad (13)$$

It is seen that Eq. (13) contains the terms with coefficients m_1, m_2, m_3, m_6, m_7 in Eq. (2). Besides, the terms with coefficients m_4, m_5 in Eq. (2) have been obtained through a Chapman–Enskog solution of the phonon Boltzmann equation within the first-order expansion [9], which results in the well-known G–K equation (4). Explorations of the kinetic foundation of generalized heat transport equations are still pending, for instance, the consideration of higher-order Chapman–Enskog expansion. On the other hand, Eq. (2) has been verified to be compatible with second law of thermodynamics in the framework of extended thermodynamics. A weakly nonlocal and nonlinear heat transport equation in a more general form than Eq. (2) was derived through the classical Lagrange multiplier method in rational extended thermodynamics [38]. In a very recent work [39], nonlinear thermodynamic force–flux relations were proposed as an extension of the usual linear ones in extended irreversible thermodynamics, resulting in the nonlinear terms in Eq. (2). Nevertheless, further efforts are still desirable for a clear physical interpretation of the nonlinear force–flux relations. The serious connection between phonon kinetic theory and generalized nonequilibrium thermodynamics remains also an open question. However, the aim in the present work is not the physical foundations (including kinetic theory and thermodynamic aspects) of unknown terms, but the physical consequences of the hitherto known terms with coefficients m_i in propagation of thermal perturbation (and, especially, heat wave propagations), as the topic in next section.

3. Heat waves around nonequilibrium steady states

The nonlinear and nonlocal terms in Eq. (2) make it impracticable to obtain a general solution of it. Here, as previously done in Refs. [26–28], Eq. (2) will be linearized around a reference nonequilibrium steady state characterized by a heat flux $\mathbf{q}_0 = (q_{x0}, 0, 0)$. In this way, the nonlinear and nonlocal terms will contribute explicitly to the speed of small thermal perturbation propagating in nonequilibrium steady states. The skipped mathematical complexities will be considered in future work. Both heat wave propagations parallel to $\mathbf{q}_0$ and orthogonal to $\mathbf{q}_0$, both for longitudinal waves and for transverse waves are thoroughly studied, as shown in Fig. 1. In practical view, these situations are easy to implement on a two-dimensional nanosystem, for instance a graphene sheet or a silicon thin layer.

For convenience, the nonlinear and nonlocal terms are classified into three categories: (i) nonlinear nonlocal terms of heat flux with coefficients: m_1, m_2, m_3 (Section 3.1); (ii) linear nonlocal terms of heat flux with coefficients: m_4, m_5 (Section 3.2); (iii) purely nonlinear terms with coefficients: m_6, m_7 (Section 3.4). Therefore, the effects of these three kinds of terms on the phase speed or front speed of different kinds of heat waves shown in Fig. 1 will be explored respectively. To obtain the phase speed or front speed of a small thermal perturbation, a perturbation method is used to linearize the heat differential equation by combining the respective simplified forms of Eq. (2) with the energy balance equation (7). In doing so, the coefficients λ, τ and m_i are taken as constants rather than temperature dependent. Such temperature dependence will lead to further nonlinear terms, which would play a role at low frequencies. Here we have focused our attention on relatively high-frequency thermal perturbations (namely: $\omega\tau \gg 1$), thus neglect this dependence. The scheme of this method will be elucidated through the solution in Section 3.1 as an example. Only the results will be provided in subsequent subsections, without the solution details anymore.

3.1. Nonlinear nonlocal terms of heat flux

Here, the situation with coefficients $m_1, m_2, m_3 \neq 0$, $m_4, m_5, m_6, m_7 = 0$ is considered. With this simplification, Eq. (2) becomes:

$$\tau\frac{\partial \mathbf{q}}{\partial t} + \mathbf{q} = -\lambda\nabla T + m_1\mathbf{q}\nabla\cdot\mathbf{q} + m_2\mathbf{q}\cdot\nabla\mathbf{q} + m_3\nabla\mathbf{q}^2. \quad (14)$$

In the reference one-dimensional nonequilibrium steady state, we have $\partial q_{x0}/\partial x = 0$ from the energy balance equation (7). Thus the initial solution of Eq. (14) becomes: $q_{x0} = -\lambda\partial T_0/\partial x$, with the subscript 0 representing the steady state. Then the perturbations of heat flux and temperature around the steady state will be studied.

3.1.1. Case I (longitudinal waves propagating along $\mathbf{q}_0$: $\delta q_x(x,t)$)

For longitudinal wave propagation along x-direction, Eq. (14) reduces to the one-dimensional form:

$$\tau\frac{\partial q_x}{\partial t} + q_x = -\lambda\frac{\partial T}{\partial x} + M_1 q_x\frac{\partial q_x}{\partial x}, \quad (15)$$

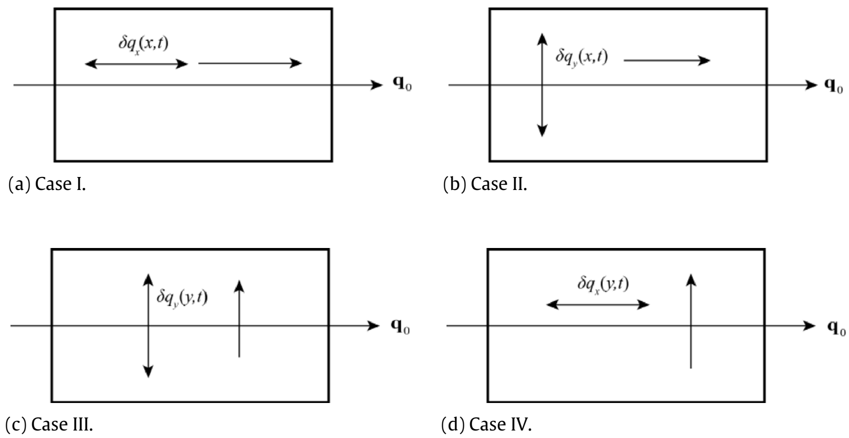
with $M_1 = m_1 + m_2 + 2m_3$. Combination of Eq. (15) with the energy balance equation (7) gives rise to a temperature differential equation with the nonlinear terms neglected:

$$\tau\frac{\partial^2 T}{\partial t^2} + \frac{\partial T}{\partial t} = \alpha\frac{\partial^2 T}{\partial x^2} + M_1 q_x\frac{\partial^2 T}{\partial x\partial t}, \quad (16)$$

where $\alpha = \lambda/C_V$ is the thermal diffusivity.

The following temperature and heat flux perturbations are considered:

$$q_x(x,t) = q_{x0} + \delta q_x(x,t), \qquad T(x,t) = T_0(x) + \delta T(x,t). \quad (17)$$



(a) Case I. (b) Case II. (c) Case III. (d) Case IV.

Fig. 1. Schematics of heat wave propagations in nonequilibrium steady state: (a) longitudinal wave propagating along x-direction, denoted as case I; (b) transverse wave propagating along x-direction, denoted as case II; (c) longitudinal wave propagating along y-direction, denoted as case III; (d) transverse wave propagating along y-direction, denoted as case IV. The double arrow represents the direction of the thermal perturbation, whereas the single arrow represents the propagating direction of heat waves.

The perturbation parts in Eq. (17) are assumed to be much smaller than the steady-state parts, such that the high-order terms in the perturbations ($\delta T\delta q_x$, $\delta T\delta T$, $\delta q_x\delta q_x$) are negligible. Putting Eq. (17) into Eq. (16) thus results in the linearized form of Eq. (16):

$$\tau\frac{\partial^2\delta T}{\partial t^2}+\frac{\partial\delta T}{\partial t}=\alpha\frac{\partial^2\delta T}{\partial x^2}+M_1q_{x0}\frac{\partial^2\delta T}{\partial x\partial t}. \tag{18}$$

For simplicity, a harmonic temperature perturbation is assumed to compute the phase speed of heat waves:

$$\delta T\left(x,t\right)=\overline{T}\exp\left[i\left(\omega t-kx\right)\right], \tag{19}$$

with $\overline{T}$, ω, k denoting separately the amplitude, frequency and wave number of the harmonic perturbation. The dispersion equation for the heat wave is derived by substituting Eq. (19) into Eq. (18):

$$k^2-\frac{M_1q_{x0}}{\alpha}\omega k-\frac{\tau}{\alpha}\omega^2+i\frac{\omega}{\alpha}=0. \tag{20}$$

The high-frequency heat waves ($\omega^2\gg\omega$) are taken into account hereafter in the present work since the phase speed in this limit yields the speed of short thermal pulse propagation. Thus Eq. (20) reduces to:

$$k^2-\frac{M_1q_{x0}}{\alpha}\omega k-\frac{\tau}{\alpha}\omega^2=0. \tag{21}$$

The solution of Eq. (21) is:

$$k=\sqrt{\frac{\tau}{\alpha}}\left(\Lambda_1\pm\sqrt{1+\Lambda_1^2}\right)\omega,\quad \text{with } \Lambda_1=\frac{M_1q_{x0}}{2\sqrt{\alpha\tau}}. \tag{22}$$

In Eq. (22), the 'plus' sign represents a positive wave number, where the heat wave propagates in the same direction with $\mathbf{q}_0$, and the 'minus' sign represents an opposite direction of propagation. Therefore, the phase speeds of heat wave in the positive and opposite directions are obtained as:

$$v_{\mathrm{p}}^{\pm}=\left|\frac{\omega}{\mathrm{Re}\left(k\right)}\right|=\sqrt{\frac{\alpha}{\tau}}\frac{1}{\sqrt{1+\Lambda_1^2}\pm\Lambda_1}, \tag{23}$$

where 'Re' denotes the real part of a complex number. Eq. (23) indicates that in nonequilibrium steady state, the heat wave propagates with different phase speeds in the direction of and in the opposite direction of the steady-state heat flux $\mathbf{q}_0$. The difference between the two phase speeds vanishes in equilibrium steady state ($q_{x0}=0$), wherein the phase speed reduces to the well-known result derived from the C–V law: $\sqrt{\alpha/\tau}$. The result Eq. (23) has been already obtained in Refs. [23,24,26–28]. In subsequent subsections we will consider the situations that have not yet been examined to our best knowledge, which will provide additional information about the coefficients in Eq. (2).

3.1.2. *Case II (transverse waves propagating along $\mathbf{q}_0$: $\delta q_y(x,t)$)*

For transverse wave propagating along x-direction, the x-component and y-component of Eq. (14) become respectively:

$$\tau\frac{\partial q_x}{\partial t}+q_x=-\lambda\frac{\partial T}{\partial x}+2m_3\delta q_y\frac{\partial\delta q_y}{\partial x}, \tag{24}$$

$$\tau\frac{\partial\delta q_y}{\partial t}+\delta q_y=-\lambda\frac{\partial T}{\partial y}+m_2q_{x0}\frac{\partial\delta q_y}{\partial x}. \tag{25}$$

Neglecting the second-order small term, combined with $q_x=q_{x0}=-\lambda\partial T_0/\partial x$, Eq. (24) vanishes. Based on the energy balance equation (7), the time derivative of temperature is zero, thus the temperature is independent of time, i.e. $T=T_0(x)$. Therefore Eq. (25) reduces to:

$$\tau\frac{\partial\delta q_y}{\partial t}+\delta q_y=m_2q_{x0}\frac{\partial\delta q_y}{\partial x}. \tag{26}$$

The solution of Eq. (26) adopts the expression of a decaying traveling signal:

$$\delta q_y\left(x,t\right)=\delta q_{y0}\exp\left(-t/\tau\right)f\left(ct\pm x\right), \tag{27}$$

where f is an arbitrary function. Putting Eq. (27) into Eq. (26) gives rise to the propagation speed of the perturbation front along the x-direction:

$$c=\pm\frac{m_2q_{x0}}{\tau}. \tag{28}$$

Eq. (28) indicates that the perturbation does not propagate in equilibrium state ($q_{x0}=0$), but simply decays with time exponentially, with relaxation time τ independent on the form of $f(x)$. Furthermore, it is inferred from Eq. (28) that the perturbation propagates only in the opposite direction of the steady-state heat flux. Since the signal decay exponentially in time, the displacement of perturbation will be small.

3.1.3. *Case III (longitudinal waves propagating orthogonal to $\mathbf{q}_0$: $\delta q_y(y,t)$)*

For longitudinal wave propagating along y-direction in principle, $\delta T(y,t)$ in the propagation could depend also on x, i.e. the perturbation becomes $\delta T(x,y,t)$, but here $\delta T(y,t)$ is simply considered for illustration. Thus, the temperature field will be $T(x,y,t) = T_0(x) + \delta T(y,t)$, whereas the heat flux field being $\mathbf{q}(y,t) = (q_{x0}, \delta q_y(y,t), 0)$.

The x-component of Eq. (14) vanishes and the y-component becomes after neglecting the higher-order terms:

$$\tau \frac{\partial \delta q_y}{\partial t} + \delta q_y = -\lambda \frac{\partial \delta T}{\partial y}. \tag{29}$$

The energy balance equation (7) reduces to:

$$C_V \frac{\partial \delta T}{\partial t} = -\frac{\partial \delta q_y}{\partial y}. \tag{30}$$

Combination of Eqs. (29) and (30) gives the differential equation of temperature perturbation:

$$\tau \frac{\partial^2 \delta T}{\partial t^2} + \frac{\partial \delta T}{\partial t} = \alpha \frac{\partial^2 \delta T}{\partial y^2}. \tag{31}$$

The phase speed of high-frequency heat wave is derived from Eq. (31) as:

$$v_{\mathrm{p}}^{\pm} = \sqrt{\frac{\alpha}{\tau}}. \tag{32}$$

Comparison of Eq. (32) to Eq. (23) shows that the steady-state heat flux $\mathbf{q}_0$ has a relevance only to the longitudinal heat wave propagating parallel to its direction. The phase speed of longitudinal heat wave propagating orthogonal to $\mathbf{q}_0$ is independent on the heat flux. Of course, actually the phase speed v_{p} may depend on temperature, thus a pulse initially orthogonal to the x-axis would become distorted, which would cause a perturbation of the x component of heat flux too. This more complicated situation needs further investigation in future work.

3.1.4. Case IV (transverse waves propagating orthogonal to $\mathbf{q}_0$: $\delta q_x(y,t)$)

For transverse wave propagating along y-direction, the temperature will not vary with time based on the energy balance equation (7). The y-component of Eq. (14) vanishes and the x-component becomes:

$$\tau \frac{\partial \delta q_x}{\partial t} + \delta q_x = 0. \tag{33}$$

The solution of Eq. (33) is explicitly:

$$\delta q_x = \delta q_{x0} \exp\left(-\frac{t}{\tau}\right). \tag{34}$$

Eq. (34) indicates that the thermal perturbation will not propagate but only decays exponentially with time. Comparison of Eq. (34) to Eq. (27) also reveals that the steady-state heat flux $\mathbf{q}_0$ has only a relevance to the transverse heat wave propagating parallel to its direction.

3.2. Linear nonlocal terms of heat flux

Here, the situation with coefficients $m_1, m_2, m_3 = 0,\ m_4, m_5 \neq 0,\ m_6, m_7 = 0$ is examined, in such a way that Eq. (2) becomes:

$$\tau \frac{\partial \mathbf{q}}{\partial t} + \mathbf{q} = -\lambda \nabla T + m_4 \nabla^2 \mathbf{q} + m_5 \nabla (\nabla \cdot \mathbf{q}). \tag{35}$$

At the reference nonequilibrium steady state, we have the solution for Eq. (35) : $q_{x0} = -\lambda \partial T_0 / \partial x$.

3.2.1. Case I (longitudinal waves propagating along $\mathbf{q}_0$: $\delta q_x(x,t)$)

For longitudinal wave propagation along x-direction, Eq. (35) reduces to the one-dimensional form:

$$\tau \frac{\partial q_x}{\partial t} + q_x = -\lambda \frac{\partial T}{\partial x} + M_2 \frac{\partial^2 q_x}{\partial x^2}, \tag{36}$$

with $M_2 = m_4 + m_5$. Combined with the energy balance equation (7), Eq. (36) gives rise to the linearized form of temperature differential equation:

$$\tau \frac{\partial^2 \delta T}{\partial t^2} + \frac{\partial \delta T}{\partial t} = \alpha \frac{\partial^2 \delta T}{\partial x^2} + M_2 \frac{\partial^3 \delta T}{\partial x^2 \partial t}. \tag{37}$$

Through similar procedures in Section 3.1.1, the phase speed of high-frequency thermal perturbation based on Eq. (37) is obtained:

$$v_{\mathrm{p}}^{\pm} = \sqrt{\frac{\alpha}{\tau}} \mathrm{Re}\left(\sqrt{1 + \Lambda_2 i}\right) \quad \text{with } \Lambda_2 = \frac{M_2 \omega}{\alpha}. \tag{38}$$

Eq. (38) indicates that in nonequilibrium steady state, the thermal perturbation propagates with the same phase speed in the direction and in the opposite direction of $\mathbf{q}_0$. Furthermore, the phase speed is independent of the value of $\mathbf{q}_0$. This is not surprising since $\mathbf{q}_0$ does not appear in the linearized heat transport equation (37), in contrast to the nonlinear one Eq. (18) in Section 3.1. However, the phase speed equation (38) increases with ω infinitely, which leads to the non-physical divergence of propagation speed of a short heat pulse in the limit of $\omega \to \infty$. This behavior results from the parabolic characteristic of Eq. (37). In Section 3.3, we provide a generalized equation for Eq. (35), which gives an upper bound for the propagation speed of thermal perturbation.

3.2.2. Case II (transverse waves propagating along $\mathbf{q}_0$: $\delta q_y(x,t)$)

For transverse wave propagating along x-direction the temperature does not vary with time based on the energy balance equation (7), and the x-component of Eq. (35) vanishes, with the y-component becoming:

$$\tau \frac{\partial \delta q_y}{\partial t} + \delta q_y = m_4 \frac{\partial^2 \delta q_y}{\partial x^2}. \tag{39}$$

The solution of Eq. (39) adopts the expression of a traveling signal:

$$\delta q_y(x,t) = \delta q_{y0} \exp(-t/\tau) \exp[\eta (ct \pm x)], \tag{40}$$

with η a constant. Substituting Eq. (40) into Eq. (39) gives rise to the front speed of the thermal perturbation:

$$c = \frac{m_4 \eta}{\tau}. \tag{41}$$

The propagation speed can be determined once the initial signal $\delta q_y(x,0)$ (or η) is given, and is independent of the steady-state heat flux. Thus the propagation speed in nonequilibrium steady state is the same as that in equilibrium state.

3.2.3. Case III (longitudinal waves propagating orthogonal to $\mathbf{q}_0$: $\delta q_y(y,t)$)

For longitudinal wave propagating along y-direction, as in Section 3.1.3, after imposing the perturbation the temperature field and heat flux become: $T(x,y,t) = T(x) + \delta T(y,t)$ and $\mathbf{q}(y,t) = (q_{x0}, \delta q_y(y,t), 0)$.

The x-component of Eq. (35) vanishes and the y-component becomes:

$$\tau \frac{\partial \delta q_y}{\partial t} + \delta q_y = -\lambda \frac{\partial \delta T}{\partial y} + M_2 \frac{\partial^2 \delta q_y}{\partial y^2}. \tag{42}$$

Combination of Eq. (42) with the energy balance equation (7) results in the linearized form of temperature differential equation:

$$\tau \frac{\partial^2 \delta T}{\partial t^2} + \frac{\partial \delta T}{\partial t} = \alpha \frac{\partial^2 \delta T}{\partial y^2} + M_2 \frac{\partial^3 \delta T}{\partial y^2 \partial t}. \tag{43}$$

Eq. (43) has an identical form to Eq. (37) except the spatial coordinate. Thus the phase speed of high-frequency thermal perturbation based on Eq. (43) is just as Eq. (38). The longitudinal heat waves have the same phase speed in parallel to and orthogonal to the steady-state heat flux.

3.2.4. Case IV (transverse waves propagating orthogonal to $\mathbf{q}_0$: $\delta q_x(y, t)$*)*

For transverse wave propagating along y-direction, the temperature will not vary with time based on the energy balance equation (7). The y-component of Eq. (35) vanishes and the x-component becomes:

$$\tau \frac{\partial \delta q_x}{\partial t} + \delta q_x = m_4 \frac{\partial^2 \delta q_x}{\partial y^2}. \tag{44}$$

Eq. (44) has an identical form to Eq. (39). Thus the front speed of the perturbation based on Eq. (44) is just as Eq. (41). The transverse heat pulses have the same front speed in parallel to and orthogonal to the steady-state heat flux.

The present Section 3.2 indicates that the steady-state heat flux has no relevance to the heat wave propagation based on the generalized heat transport equation Eq. (35) with linear nonlocal terms.

3.3. A generalized G–K model

It has been seen in Sections 3.2.1 and 3.2.3 that the Eq. (35), which corresponds to the G–K equation (4), results in a thermal perturbation propagation with a nonphysical infinite phase speed. To remedy this anomaly, a generalized G–K equation was proposed in the frame of extended irreversible thermodynamics [40,41]:

$$\begin{aligned} &\tau \frac{\partial \mathbf{q}}{\partial t} + \mathbf{q} = -\lambda \nabla T - \nabla \cdot \mathbf{Q} \\ &\tau_2 \frac{\partial \mathbf{Q}}{\partial t} + \mathbf{Q} = -\lambda_2 \nabla \mathbf{q}, \end{aligned} \tag{45}$$

where $\mathbf{Q}$ is the flux of heat flux, and τ_2 is the relaxation time of $\mathbf{Q}$. Combination of Eq. (45) and the energy balance equation (7) results in the temperature differential equation:

$$\begin{aligned} &\tau\tau_2 \frac{\partial^3 T}{\partial t^3} + (\tau + \tau_2) \frac{\partial^2 T}{\partial t^2} + \frac{\partial T}{\partial t} \\ &= \alpha \nabla^2 T + (\alpha \tau_2 + \lambda_2) \frac{\partial}{\partial t} \left(\nabla^2 T\right). \end{aligned} \tag{46}$$

Heat wave propagation of case I is merely considered to illustrate the main features of the generalized G–K model Eq. (45), (46). Here Case II–IV is not taken into account to avoid repeat. Through similar procedures, the phase speed of high-frequency heat wave is obtained:

$$v_{\mathrm{p}}^{\pm} = \sqrt{\frac{\alpha}{\tau}} \mathrm{Re} \left(\sqrt{1 + \frac{\lambda_2}{\tau_2 \alpha} - \frac{1}{\tau_2 \omega} i} \right). \tag{47}$$

In comparison to Eq. (38) based on the G–K model, Eq. (47) based on the generalized G–K model results in a finite phase speed ($v_{\mathrm{p}}^{\pm} = \sqrt{\frac{\alpha}{\tau}} \sqrt{1 + \frac{\lambda_2}{\tau_2 \alpha}}$) of heat pulse propagation in the limit of $\omega \to \infty$. Therefore the generalized G–K equation eliminates the paradox of infinite speed of thermal perturbation propagation.

3.4. Purely nonlinear terms of heat flux

Here, the situation with coefficients $m_1, m_2, m_3 = 0$, $m_4, m_5 = 0$, $m_6, m_7 \neq 0$ is explored. With this simplification, Eq. (2) becomes:

$$\tau \frac{\partial \mathbf{q}}{\partial t} + \mathbf{q} = -\lambda \nabla T + m_6 \mathbf{q} (\mathbf{q} \cdot \nabla T) + m_7 \mathbf{q}^2 \nabla T. \tag{48}$$

At the reference nonequilibrium steady state, we have the solution for Eq. (48): $q_{x0} = -\lambda_{\mathrm{eff}} \partial T_0 / \partial x$, with the effective thermal conductivity: $\lambda_{\mathrm{eff}} = \lambda - (m_6 + m_7) q_{x0}^2$. In more general view, Eq. (48) could be treated as a generalized C–V equation with an effective thermal conductivity tensor as: $\boldsymbol{\lambda} = \lambda \mathbf{I} - m_6 \mathbf{q}\mathbf{q} - m_7 \mathbf{q}^2 \mathbf{I}$, which exactly reduces to λ_{eff} in one-dimensional situation

3.4.1. Case I (longitudinal waves propagating along $\mathbf{q}_0$: $\delta q_x(x, t)$*)*

For longitudinal wave propagation along x-direction, Eq. (48) reduces to the one-dimensional form:

$$\tau \frac{\partial q_x}{\partial t} + q_x = -\lambda \frac{\partial T}{\partial x} + (m_6 + m_7) q_x^2 \frac{\partial T}{\partial x}. \tag{49}$$

Combined with the energy balance equation (7), Eq. (49) gives rise to the linearized temperature differential equation:

$$\tau \frac{\partial^2 \delta T}{\partial t^2} + \left[1 + \frac{2(m_6 + m_7)}{\lambda_{\mathrm{eff}}} q_{x0}^2\right] \frac{\partial \delta T}{\partial t} = \alpha_{\mathrm{eff}} \frac{\partial^2 \delta T}{\partial x^2}, \tag{50}$$

with the effective diffusivity defined as: $\alpha_{\mathrm{eff}} = \lambda_{\mathrm{eff}} / C_V$.

The phase speed of high-frequency heat wave based on Eq. (50) is thus obtained:

$$v_{\mathrm{p}}^{\pm} = \sqrt{\frac{\alpha_{\mathrm{eff}}}{\tau}}. \tag{51}$$

Eq. (51) has an identical form to the phase speed $\sqrt{\alpha/\tau}$ based on the C–V equation. This could be understood since Eq. (48) is a generalized C–V equation with the effective thermal conductivity λ_{eff}.

3.4.2. Case II (transverse waves propagating along $\mathbf{q}_0$: $\delta q_y(x, t)$*)*

For transverse wave propagating along x-direction, the temperature does not vary with time based on the energy balance equation (7), and the x-component of Eq. (48) vanishes, whereas the y-component reduces to:

$$\tau \frac{\partial \delta q_y}{\partial t} + \frac{\lambda_{\mathrm{eff},1}}{\lambda_{\mathrm{eff}}} \delta q_y = 0, \tag{52}$$

with $\lambda_{\mathrm{eff},1} = \lambda - m_7 q_{x0}^2$. The solution of Eq. (52) is explicitly:

$$\delta q_y(x, t) = \delta q_{y0} \exp\left(-\frac{\lambda_{\mathrm{eff},1}}{\lambda_{\mathrm{eff}}} t/\tau\right). \tag{53}$$

Eq. (53) shows that the heat pulse perturbation will not propagate but only decay exponentially with time. Based on the identification of the coefficients in Table 1, $m_6 > 0$, thus $\lambda_{\mathrm{eff},1} > \lambda_{\mathrm{eff}}$ inferring the decay rate of perturbation around nonequilibrium steady state is larger than that around equilibrium state.

3.4.3. Case III (longitudinal waves propagating orthogonal to $\mathbf{q}_0$: $\delta q_y(y, t)$*)*

For longitudinal wave propagating along y-direction, as in Sections 3.1.3 and 3.2.3, after imposing the perturbation, the temperature and heat flux fields become: $T(x, y, t) = T(x) + \delta T(y, t)$ and $\mathbf{q}(y, t) = (q_{x0}, \delta q_y(y, t), 0)$.

The x-component of Eq. (48) vanishes and the y-component becomes:

$$\tau \frac{\partial \delta q_y}{\partial t} + \left(1 - m_6 q_{x0} \frac{\partial T_0}{\partial x}\right) \delta q_y = -\lambda_{\mathrm{eff},1} \frac{\partial \delta T}{\partial y}. \tag{54}$$

Combination of Eq. (54) with the energy balance equation (7) results in the linearized form of temperature differential equation:

$$\tau \frac{\partial^2 \delta T}{\partial t^2} + \frac{\lambda_{\mathrm{eff},1}}{\lambda_{\mathrm{eff}}} \frac{\partial \delta T}{\partial t} = \alpha_{\mathrm{eff},1} \frac{\partial^2 \delta T}{\partial y^2}, \tag{55}$$

with the effective diffusivity defined as: $\alpha_{\mathrm{eff},1} = \lambda_{\mathrm{eff},1} / C_V$.

The phase speed of high-frequency heat wave based on Eq. (55) is thus obtained:

$$v_{\mathrm{p}}^{\pm} = \sqrt{\frac{\alpha_{\mathrm{eff},1}}{\tau}}. \tag{56}$$

Both Eqs. (51) and (56) indicate that the steady-state heat flux has a relevance to the longitudinal heat wave propagation based on Eq. (48). The relevance is slightly different in the propagating direction parallel to and orthogonal to the heat flux $\mathbf{q}_0$.

Table 2
Heat wave propagations in nonequilibrium steady states characterized by an average heat flux $\mathbf{q}_0$ along the positive direction of x-axis.

Cases	A generalized heat transport equation (Eq. (2))			A generalized G–K model
	$m_1, m_2, m_3 \neq 0$ (Eq. (14))	$m_6, m_7 \neq 0$ (Eq. (48))	$m_4, m_5 \neq 0$ (Eq. (35))	Eq. (45)
I $\delta q_x(x,t)$	$v_p^{\pm} = \sqrt{\frac{\alpha}{\tau}} \frac{1}{\sqrt{1+\Lambda_1^2} \pm \Lambda_1}$	$v_p^{\pm} = \sqrt{\frac{\alpha_{\text{eff}}}{\tau}}$	$v_p^{\pm} = \sqrt{\alpha/\tau} \times \text{Re}\left(\sqrt{1+\Lambda_2 i}\right)$	$v_p^{\pm} = \sqrt{\frac{\alpha}{\tau}}\sqrt{1+\frac{\lambda_2}{\tau_2 \alpha}}$
II $\delta q_y(x,t)$	$c = \pm \frac{m_2 q_{x0}}{\tau}$	Don't propagate	$c = \frac{m_4 \eta}{\tau}$	–
III $\delta q_y(y,t)$	$v_p^{\pm} = \sqrt{\frac{\alpha}{\tau}}$	$v_p^{\pm} = \sqrt{\frac{\alpha_{\text{eff},1}}{\tau}}$	Same as I	–
IV $\delta q_x(y,t)$	Don't propagate	Don't propagate	Same as II	–

Notes:
1. $\Lambda_1 = \frac{M_1 q_{x0}}{2\sqrt{\alpha\tau}}$, $\Lambda_2 = \frac{M_2 \omega}{\alpha}$, with $M_1 = m_1 + m_2 + 2m_3$, $M_2 = m_4 + m_5$, thermal diffusivity $\alpha = \lambda / C_V$;
2. Effective thermal conductivity: $\lambda_{\text{eff}} = \lambda - (m_6 + m_7) q_{x0}^2$, $\lambda_{\text{eff},1} = \lambda - m_7 q_{x0}^2$;
3. Effective thermal diffusivity: $\alpha_{\text{eff}} = \lambda_{\text{eff}} / C_V$, $\alpha_{\text{eff},1} = \lambda_{\text{eff},1} / C_V$.

3.4.4. Case IV (transverse waves propagating orthogonal to $\mathbf{q}_0$: $\delta q_x(y,t)$)

For transverse wave propagating along y-direction, the temperature will not vary with time based on the energy balance equation (7). The y-component of Eq. (48) vanishes and the x-component becomes:

$$\tau \frac{\partial \delta q_x}{\partial t} + \left(\frac{2\lambda}{\lambda_{\text{eff}}} - 1\right) \delta q_x = 0. \tag{57}$$

The solution of Eq. (57) is explicitly:

$$\delta q_x(y,t) = \delta q_{x0} \exp\left[-\left(\frac{2\lambda}{\lambda_{\text{eff}}} - 1\right) t/\tau\right]. \tag{58}$$

Eq. (58) indicates that the heat pulse perturbation will not propagate but only decay exponentially with time. Both Eqs. (53) and (58) reveal that the steady-state heat flux has a relevance to the decay rates of transverse heat pulse imposed both parallel and orthogonal to its direction.

4. Discussions

The ultimate aim of the present work is to explore further how heat waves could contribute to the measurement of coefficients appearing in Eq. (2). This is not an easy task due to the number of coefficients appearing in Eq. (2), whose measurement requires several different kinds of independent experiments. First of all, the specific heat capacity C_V can be got from equilibrium measurements, whereas the thermal conductivity λ can be derived from near-equilibrium steady-state measurements. A measurement of the phase speed of heat wave around equilibrium states will give the relaxation time τ. Thus the coefficients m_i remain to be measured. In principle, one-dimensional steady-state measurement (implying $\partial q_{x0}/\partial x = 0$) would allow to obtain $m_6 + m_7$, since in this case Eq. (2) reduces to:

$$q_{x0} = -\left[\lambda - (m_6 + m_7) q_{x0}^2\right] \frac{\partial T_0}{\partial x}. \tag{59}$$

To discriminate between m_6 and m_7, a two-dimensional steady-state heat conduction experiment should be designed, considering a homogeneous heat flux q_0 along the x axis, the relation between q_y and $\partial T/\partial y$ along the y-axis being:

$$q_y = -\frac{\lambda - (m_6 + m_7) q_y^2}{1 - m_6 q_{x0} \partial T_0/\partial x} \frac{\partial T}{\partial y}. \tag{60}$$

For the case of small values of q_y, one would have:

$$q_y \approx -\frac{\lambda}{1 - m_6 q_{x0} \partial T_0/\partial x} \frac{\partial T}{\partial y}, \tag{61}$$

in such a way that m_6 could be obtained. Steady-state measurements in radial heat transport, where the steady-state radial heat flux adopts the form $q_r(r) = 1/r$, could provide information on coefficients $m_1 - m_5$.

But here we focus on dynamical measurements of m_1–m_7, based on heat wave propagation along non-equilibrium steady states. The results of the analysis in Section 3 are summarized in Table 2. Through the experimental measurements, it can be also determined which terms dominate in the generalized heat transport equation. In this way, the present work would contribute to the fundamental understanding and macroscopic modeling of the heat transport in nanosystems and in ultrafast pulse heating processes.

In the present analysis discussed in Section 3 the linear nonlocal terms (m_4, m_5), the nonlinear nonlocal terms (m_1, m_2, m_3) and the purely nonlinear terms (m_6, m_7) have been accounted separately. In actual thermal transport experiments, the mixed features from all these terms may appear. However, the combination of different measurements in equilibrium and nonequilibrium situations allows for discrimination between the several coefficients. This has been the reason to consider heat wave propagation in nonequilibrium steady states.

5. Conclusions

In this work, a generalized heat transport equation including nonlinear, nonlocal and relaxation terms is proposed, which sums up diverse macroscopic models for nanoscale heat transport as special cases. Heat wave propagations in nonequilibrium steady state characterized by a heat flux $\mathbf{q}_0$ have been systematically studied and the following points are concluded:

(1) Nonlinear terms (including the nonlinear nonlocal terms and the purely nonlinear terms) make the phase speed of heat waves in nonequilibrium steady state different from that in equilibrium state. Furthermore, the phase speed along $\mathbf{q}_0$ is predicted different from that in the opposite direction of $\mathbf{q}_0$.
(2) The nonlinear nonlocal terms result in different phase or front speeds propagating parallel to and orthogonal to $\mathbf{q}_0$ for both longitudinal and transverse heat waves; in contrast, the linear nonlocal terms result in the same speeds for both kinds of waves. The purely nonlinear terms give rise to different phase speeds propagating parallel to and orthogonal to $\mathbf{q}_0$ for longitudinal heat waves, and different decay rates for transverse heat pulses.
(3) In high-frequency limit, the Guyer–Krumhansl model will result in an infinite phase speed of thermal perturbation, which is reduced to a finite value in the generalized Guyer–Krumhansl model by considering the relaxation of the flux of heat flux.

Therefore, credible experimental measurements could be designed in equilibrium and nonequilibrium states to specify the coefficients in the generalized heat transport equation (2), which would foster deeper understanding and macroscopic modeling of nanoscale heat transport. The next task could be to examine some of the genuine nonlinear effects in wave propagation. Although a topic analogous to this has been much studied in electromagnetic waves, it has been not yet received attention for heat waves. This exploration is truly needed to explore the whole possibilities of thermal waves which could probably lead to phenomena such as self-focusing or flux-limited behaviors [42]. From the mathematical point of view, these phenomena should be related to the coefficients m_i considered in this article. Exploration of Eq. (2) in other geometries, as for instance in two-dimensional heat flow in thin layers or graphene sheets [43], could also bring additional information about the influence of the non-classical terms.

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