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# Fractional Boltzmann transport equation for anomalous heat transport and divergent thermal conductivity

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ABSTRACT

Anomalous heat transport and divergent thermal conductivity have attracted increasing attention in recent years. The linearized Boltzmann transport equation (BTE) proposed by Goychuk is discussed in superdiffusive and ballistic heat conduction, which is characterized by super-linear growth of the mean-square displacement (MSD) $\langle \Delta \mathbf{x}^2 \rangle$, namely, $\langle \Delta \mathbf{x}^2 \rangle \sim t^\gamma$ with $1 < \gamma \leqslant 2$. We show that this fractional-order BTE predicts a fractional-order constitutive equation and divergent effective thermal conductivity $\kappa_{eff}$. In the long-time limit, the divergence obeys a power-law type $\kappa_{eff} \sim t^\alpha$, while the asymptotics of $\langle \Delta \mathbf{x}^2 \rangle$ reads $\gamma = \alpha + 1$. This connection between $\kappa_{eff}$ and $\langle \Delta \mathbf{x}^2 \rangle$ coincides with previous investigations such as the linear response and Lévy-walk model. The constitutive equation from Goychuk's model is compared with a class of fractional-order models termed generalized Cattaneo equation (GCE). We show that Goychuk's model is more appropriate than other models of the GCE class to describe superdiffusive and ballistic heat conduction.



## 1. Introduction

Linearized Boltzmann transport equation (BTE) [1–10] is widely applied to transport phenomena such as diffusion and heat conduction. At the macroscale, heat conduction is commonly described by classical Fourier's law, namely,

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

wherein $\mathbf{q} = \mathbf{q}(\mathbf{x}, t)$ is the heat flux, $T = T(\mathbf{x}, t)$ is the local temperature and $\kappa$ denotes the thermal conductivity. As a material property, $\kappa$ can be calculated from the linearized BTE. In kinetics, the standard Bhatnagar-Gross-Krook (BGK) model predicts

$$\kappa = \frac{5k_B^2 T \rho \tau}{2m^2} \tag{2}$$

where $\tau$ is the relaxation time, $\rho$ is the density, $m$ is the mass of one particle and $k_B$ is the Boltzmann constant. For phonon heat transport, one can obtain the following expression from the single mode relaxation time approximation [11],

$$\kappa = \frac{1}{3} |\mathbf{v}_g|^2 c \tau \tag{3}$$

with $\mathbf{v}_g$ denoting the phonon group velocity and $c$ the specific heat capacity per unit volume. When the characteristic size $L_C$ and time $\tau_C$ are comparable to (or even smaller than) the mean free path (MFP) $l$ and relaxation time $\tau$ of heat carriers, respectively, the validity of Fourier's law becomes debatable [2,12–14]. For instance, the single mode relaxation time approximation gives rise to the Cattaneo equation [14] rather than Fourier's law in non-stationary heat conduction, namely,

$$\mathbf{q} + \tau \partial_t \mathbf{q} = -\kappa \nabla T. \tag{4}$$

If $|\partial_t \mathbf{q}|$ is sufficiently large, violation of Fourier's law will emerge. In the case of constant material properties, the Cattaneo equation leads to a hyperbolic heat conduction equation of $T$ as follows

$$\partial_t T + \tau \partial_t^2 T = D \nabla^2 T, \tag{5}$$

with $D = \kappa / c$ denoting the thermal diffusivity. The hyperbolic heat equation can overcome the infinite speed of heat propagation traceable to Fourier's law, yet paired with possible unsatisfactory behaviors such as negative entropy generation [15–17] and absolute temperature [18,19]. Chen [5] discussed the single mode relaxation time approximation based on a more physically meaningful approach, which divides the distribution function into ballistic and diffusive parts. Then, a calculation for the ballistic heat flux and a hyperbolic equation for the diffusive temperature distribution arise, which are termed as ballistic-diffusive heat conduction

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equations. One inconsistency induced by the Cattaneo and ballistic-diffusive heat equations is the artificial wave front in the diffusive regime. In order to avoid this inconsistency, Razi-Naqvi and Waldenstrøm [20] suggested a new heat equation with a time-dependent effective thermal diffusivity, which likewise guarantees a finite speed of heat propagation. This model can also be derived from a linearized BTE by assuming Gaussian phonon radiative transfer. More accurate models can emerge from more refined BTEs. Callaway's dual relaxation approximation [21] is a typical one, which provides better estimations for the thermal conductivity of two-dimensional materials than the single mode relaxation time approximation [22]. Besides relaxation, Callaway's model also predicts a non-Fourier effect named nonlocality [2], which reflect spatial derivatives like $\nabla(\nabla\cdot\mathbf{q})$ and $\nabla^2\mathbf{q}$.

Another remarkable non-Fourier behavior is the length-dependence (or time-dependence) of the effective thermal conductivity $\kappa_{eff}$ [23–28], which is usually termed as anomalous heat conduction. In anomalous heat conduction, $\kappa_{eff}$ is commonly defined as

$$\kappa_{eff} = -\frac{q_{1D}L_C}{\Delta T}, \tag{6}$$

where $q_{1D}$ is the one-dimensional (1D) heat flux and $\Delta T$ is the temperature difference. Most existing investigations [4,29–31] based on linearized BTEs predict convergence in the limit $L_C \to +\infty$, i.e.,

$$\kappa_{eff} = \frac{1}{1+CKn}\kappa_0 \tag{7}$$

In Eq. (7), $Kn = l/L_C$ stands for the Knudsen number, $C$ can be calculated theoretically or numerically, and $\kappa_0$ generally equals to the bulk thermal conductivity. Nevertheless, divergence like $\lim_{L_C\to+\infty}\kappa_{eff} = +\infty$ has been widely observed in low-dimensional systems. In 1D or quasi-one-dimensional systems, the divergence theoretically obeys a power-law type $\kappa_{eff} \sim L_C^\beta$ (or $\kappa_{eff} \sim t^\beta$) within $0<\beta\leqslant 1$, while in two-dimensional (2D) cases, the divergence commonly becomes logarithmic, $\kappa_{eff} \sim \ln\ L_C$ (or $\kappa_{eff} \sim \ln\ t$). Low-dimensional momentum-conserving systems [27] are typical examples. Based on fluctuating hydrodynamics and the renormalization group theory, Narayan and Ramaswamy [27] deduced $\beta = 1/3$ for 1D momentum-conserving systems, while in 2D cases, the logarithmic divergence occurs. The Fermi-Pasta-Ulam (FPU) model is another classical class, where the mode-coupling theory [24] predicts $\beta = 2/5$ in 1D cases and $\kappa_{eff} \sim \ln\ L_C$ in 2D cases. Such divergence also emerges from the linearized BTE class in phonon heat transport. For instance, one can recover $\kappa_{eff} \sim L_C^\beta$ through assuming the following mode-dependent relaxation time [23]

$$\tau = \tau(q) \sim q^a, \quad a = \frac{1}{\beta-1} < -1, \tag{8}$$

with $q$ the phonon mode, while $\kappa_{eff} \sim \ln\ L_C$ emerges from $a=-1$. Renormalized phonons and effective phonon theory [32] can give rise to $\kappa_{eff} \sim L_C^\beta$ as well, which introduces a weight factor $P = P(q)$ in the conventional Deybe formula,

$$\kappa_{eff} = \frac{c}{2\pi}\int_0^{2\pi}\left|\mathbf{v}_g(q)\right|^2\tau(q)P(q)dq, \tag{9}$$

The weight factor should fulfill the normalization condition $\int_0^{2\pi}P(q)dq = 2\pi$, and $\kappa_{eff} \sim L_C^\beta$ arises from $P(q) \sim q^{-\beta}$ in the long wave-length limit $q \to 0$.

In anomalous heat conduction, the length-dependence of $\kappa_{eff}$ is usually connected to the long-time asymptotics of the mean-square displacement (MSD), $\langle\Delta\mathbf{x}^2\rangle \sim t^\gamma$ [25,26,33,34]. Relying on the Lévy-walk model, Denisov *et al.* [33] found $\beta = \gamma - 1$, which occurs from the Green-Kubo formula as well [25]. Li and Wang [26] have obtained a different scaling relation, $\beta = 2 - 2/\gamma$, which is based on the length-dependence of the mean first passage time (MFPT) in billiard gas channel models. For both $\beta = \gamma - 1$ and $\beta = 2 - 2/\gamma$, it is obvious that superdiffusive and ballistic regimes within $1<\gamma\leqslant 2$ corresponds to divergent $\kappa_{eff}$. One unsatisfactory aspect of the linearized BTE class is that such connection between the MSD and $\kappa_{eff}$ has not been established based on the linearized BTE. Therefore, the relation between divergent $\kappa_{eff}$ and superdiffusive-ballistic heat conduction remains an open issue in the framework of the linearized BTE.

In the present work, we will address this issue based on a fractional-order BTE proposed by Goychuk [35]. This phenomenological model introduced a fractional-order BGK collision term in the kinetic equation, which is applied to anomalous dielectric response in the absence of retardation effects. Here, we use this fractional-order BTE to study superdiffusive and ballistic heat conduction. We show that this fractional-order generalization predicts a fractional-order constitutive equation and divergent $\kappa_{eff}$. The divergence is then connected to the MSD, which coincides with previous investigations. The constitutive equation from this BTE is also compared with an existing class of fractional-order models termed generalized Cattaneo equation (GCE) [36].

## 2. Anomalous behaviors in heat transport

### 2.1. Kinetic heat transport

We start from the following linearized BTE

$$\partial_t f_K + \mathbf{v}\cdot\nabla f_K = \tau^\alpha D_t^\alpha\left(\frac{f_{MB}-f_K}{\tau}\right), \tag{10}$$

where $0<\alpha\leqslant 1$, $f_K = f_K(\mathbf{x}, t, \mathbf{v})$ is the single-particle distribution function, $\mathbf{v}$ is the particle velocity, $D_t^\alpha$ is the Riemann-Liouville (RL) operator and $f_{MB}$ stands for the Maxwell-Boltzmann distribution. Rigorously speaking, Eq. (10) does not have a stationary solution in non-equilibrium cases. Because $\tau^{\alpha-1}D_t^\alpha(f_{MB}-f_K)$ is nonzero unless $f_{MB} = f_K$, time-independent and non-equilibrium $f_K$ can never fulfill Eq. (10). Thus, we consider quasi-stationary heat conduction in a time-independent temperature field. In kinetics, the energy density $e = e(\mathbf{x}, t)$ and heat flux take the following forms, respectively,

$$e = \frac{3\rho k_B}{2m}T = \frac{m}{2}\int|\mathbf{v}-\mathbf{c}|^2 f_K d\mathbf{v}, \tag{11a}$$

$$\mathbf{q} = \frac{m}{2}\int|\mathbf{v}-\mathbf{c}|^2(\mathbf{v}-\mathbf{c})f_K d\mathbf{v}, \tag{11b}$$

with $\mathbf{c} = \int\mathbf{v}f_K d\mathbf{v}$. Substituting Eq. (10) into $\tau^{\alpha-1}D_t^\alpha\mathbf{q}$ yields

$$\begin{aligned}
\tau^{\alpha-1}D_t^\alpha\mathbf{q} &= \tfrac{m}{2}\int|\mathbf{v}-\mathbf{c}|^2(\mathbf{v}-\mathbf{c})\tau^{\alpha-1}D_t^\alpha f_K d\mathbf{v}\\
&= \tfrac{m}{2}\int|\mathbf{v}-\mathbf{c}|^2(\mathbf{v}-\mathbf{c})\left(\tau^{\alpha-1}D_t^\alpha f_{MB} - \mathbf{v}\cdot\nabla f_K - \partial_t f_K\right)d\mathbf{v}\\
&= -\tfrac{m}{2}\int|\mathbf{v}-\mathbf{c}|^2(\mathbf{v}-\mathbf{c})\mathbf{v}\cdot\nabla f_K d\mathbf{v} - \tfrac{m}{2}\int|\mathbf{v}-\mathbf{c}|^2(\mathbf{v}-\mathbf{c})\partial_t f_K d\mathbf{v}\\
&= -\tfrac{m}{2}\int|\mathbf{v}-\mathbf{c}|^2(\mathbf{v}-\mathbf{c})\mathbf{v}\cdot\nabla f_K d\mathbf{v} - \partial_t\mathbf{q}.
\end{aligned} \tag{12a}$$

According to the local equilibrium assumption, one can write $\nabla f_K \approx \nabla f_{MB}$ and

$$\begin{aligned}
\tau^{\alpha-1}D_t^\alpha\mathbf{q} + \partial_t\mathbf{q} &= -\tfrac{m}{2}\int|\mathbf{v}-\mathbf{c}|^2(\mathbf{v}-\mathbf{c})\mathbf{v}\cdot\nabla f_K d\mathbf{v}\\
&= -\left[\tfrac{m}{2}\int|\mathbf{v}-\mathbf{c}|^2(\mathbf{v}-\mathbf{c})\mathbf{v}\cdot\nabla f_{MB}d\mathbf{v}\right].
\end{aligned} \tag{12b}$$

For pure heat conduction problems, $\mathbf{c} = \mathbf{0}$ and $\nabla\cdot\mathbf{v} = \mathbf{0}$, and thereupon, Eq. (12b) becomes

$$\begin{aligned}\tau^{\alpha-1}D_t^\alpha\mathbf{q}+\partial_t\mathbf{q}&=-\left[\tfrac{m}{2}\int|\mathbf{v}|^2\mathbf{v}\mathbf{v}\cdot(\partial_T f_{MB})d\mathbf{v}\right]\nabla T.\\ &\Rightarrow\tau^\alpha D_t^\alpha\mathbf{q}+\tau\partial_t\mathbf{q}=-\kappa\nabla T.\end{aligned}\tag{13}$$

In Eq. (13), time-independent $T$ will lead to

$$\begin{aligned}\mathbf{q}=&\left[\left(\mathbf{q}+\tau^{\alpha-1}D_t^{\alpha-1}\mathbf{q}\right)\Big|_{t=0}\right]E_{1-\alpha,1}\left[-\left(\frac{t}{\tau}\right)^{1-\alpha}\right]\\ &-\kappa\left(\frac{t}{\tau}\right)E_{1-\alpha,2}\left[-\left(\frac{t}{\tau}\right)^{1-\alpha}\right]\nabla T,\end{aligned}\tag{14}$$

where $E_{a,b}(z)=\sum_{k=0}^{+\infty}\frac{z^k}{\Gamma(b+ak)}$ is the Mittag-Leffler function. From Eq. (14), we can find that the heat flux consists of two parts: $\mathbf{q}=\mathbf{q}_0+\mathbf{q}_T$. The first part $\mathbf{q}_0$ reflects the initial effects:

$$\mathbf{q}_0=\left[\left(\mathbf{q}+\tau^{\alpha-1}D_t^{\alpha-1}\mathbf{q}\right)\Big|_{t=0}\right]E_{1-\alpha}\left[-\left(\frac{t}{\tau}\right)^{1-\alpha}\right],\tag{15}$$

while the second part $\mathbf{q}_T$ is induced by the temperature gradient:

$$\mathbf{q}_T=-\kappa\left(\frac{t}{\tau}\right)E_{1-\alpha,2}\left[-\left(\frac{t}{\tau}\right)^{1-\alpha}\right]\nabla T.\tag{16}$$

In the long-time limit, $\mathbf{q}_0$ is asymptotic to $\mathbf{q}_0\sim t^{\alpha-1}$ and $\mathbf{q}_T$ reads $\mathbf{q}_T\sim t^\alpha$. Hence the long-time asymptotics of $\mathbf{q}$ is dominated by the temperature gradient: $\mathbf{q}\sim t^\alpha\nabla T$, which means $\kappa_{eff}\propto t^\alpha$. According to Goychuk's results [35], the MSD fulfills the following asymptotics:

$$\langle\Delta\mathbf{x}^2\rangle\sim\begin{cases}t^2, t\ll\tau\\ t^{\alpha+1}, t\gg\tau\end{cases}\tag{17}$$

Hence, one can establish a connection between the divergent exponent of $\kappa_{eff}$ and the asymptotic exponent of $\langle\Delta\mathbf{x}^2\rangle$: $\beta=\gamma-1$. One typical treatment for the time-dependence like $\kappa_{eff}\sim t^\beta$ is introducing a cut-off by the "transit time", $t\sim L_C/v_S$, where $v_S$ is the sound velocity. Then, the time-dependence $\kappa_{eff}\sim t^\beta$ is transformed into the length-dependence $\kappa_{eff}\sim L_C^\beta$. Such treatment is rather common yet an approximate estimation, which may be inapplicable in some cases. In a recent work [37], a violation of $t\propto L_C$ was found in 1D uniformly charged systems with transverse motions, which emerges from the absence of a sound wave, and the cut-off time is given by $t\propto L_C^\mu$ with $\mu=1.5\pm0.001$.

The scaling relation $\beta=\gamma-1$ has been observed in existing theoretical and computational investigations. It was first derived from the Lévy-walk model in a 1D dynamical channel [33]. A more universal deduction emerges from the linear response theory [25], which only relies on the standard energy continuity equation as follows

$$\partial_t e=-\nabla\cdot\mathbf{q},\tag{18}$$

Eq. (18) will gives rise to the following relation between the total heat current correlation function $C_{qq}(t)$ and MSD

$$\frac{d^2\langle\Delta\mathbf{x}^2\rangle}{dt^2}\propto C_{qq}(t).\tag{19}$$

In the conventional Green-Kubo approach, $\kappa_{eff}$ is formulated as

$$\kappa_{eff}=\kappa_{eff}(t)\propto\int_0^t C_{qq}(\xi)d\xi,\tag{20}$$

and one can arrive at $\beta=\gamma-1$ immediately. Recently, a computational study [38] in a single polymer chain of poly-3,4-ethylenedioxythiophene (PEDOT) supported $\beta=\gamma-1$, which uses a more realistic model potential than previous investigations and implies a possible existence of $\beta=0.5$. In a recent work by Xu and Wang [39], Eq. (19) is generalized into the following modified form

$$\frac{d^2\langle\Delta\mathbf{x}^2\rangle}{dt^2}\propto\left[C_{qq}(t)+\frac{\mathscr{P}^2}{T}\right],\tag{21}$$

where $\mathscr{P}=\mathscr{P}(t)$ is the temperature pressure. In Eq. (21), the correlation function depends on not only the MSD but also the temperature pressure. It means possible cases wherein $\beta=\gamma-1$ no longer holds, and hence, the linearized BTE is invalid in these cases likewise. Moreover, one can apply a velocity-dependent collision term [6] to Eq. (10), namely,

$$\tau=\tau(|\mathbf{v}-\mathbf{c}|)\propto|\mathbf{v}-\mathbf{c}|^\theta\tag{22}$$

This velocity-dependence will lead to a power-law temperature-dependence [6], namely, $\kappa_{eff}\propto T^{\frac{2-\theta}{2}}$. Thereafter, Eq. (10) with the velocity-dependent collision term may be used to describe heat conduction where the effective thermal conductivity satisfies $\kappa_{eff}=\kappa_{eff}(T,L_C)\propto T^\delta L_C^\beta$.

### 2.2. Phonon heat transport

In phonon heat transport, the Boltzmann equation should be reformed as

$$\partial_t f_P+\mathbf{v}_g\cdot\nabla f_P=\tau^\alpha D_t^\alpha\left(\frac{f_0-f_P}{\tau}\right),\tag{23}$$

wherein $f_P=f_P(\mathbf{x},t,\mathbf{k})$ is the phonon distribution function, $\mathbf{k}$ denotes the wave vector, $f_0=\frac{1}{\exp(\hbar\omega/k_BT)-1}$ is the Planck distribution, $\hbar$ is the reduced Planck constant, and $\omega$ is the angle frequency. The energy density of phonons is given by $e=\int f_P\hbar\omega d\mathbf{k}$, while the heat flux reads $\mathbf{q}=\int\mathbf{v}_g f_P\hbar\omega d\mathbf{k}$. Upon multiplying Eq. (23) by $\mathbf{v}_g\hbar\omega$ and integrating it over the wave vector space, we acquire

$$\partial_t\mathbf{q}+\int\hbar\omega\mathbf{v}_g\mathbf{v}_g\cdot\nabla f_P d\mathbf{k}=-\tau^{\alpha-1}D_t^\alpha\mathbf{q}.\tag{24}$$

If local equilibrium is achieved ($\nabla f_P\approx\nabla f_0$), Eq. (24) will become Eq. (13). Therefore, phonon heat transport will perform the same divergence of $\kappa_{eff}$ as kinetic heat transport in the presence of local equilibrium. For the cases far from equilibrium, the local temperature cannot be defined in the sense of local equilibrium. Chen [5] suggested to define the local temperature as a measure of the energy density, namely, $e=cT=\int\hbar\omega f_P d\mathbf{k}$. Then, $\int\hbar\omega\nabla f_P d\mathbf{k}=c\nabla T$ and Eq. (13) still holds. $\kappa_{eff}\sim L_C^\alpha$ ignores the non-Fourier behavior by $\alpha=0$, which is induced by the boundary effects [28]. Although the boundary effects generally give rise to convergence to $\kappa_{bulk}$ in existing studies, their contributions to $\kappa_{eff}$ have not been studied for $\alpha\neq0$. Only if these contributions converge or diverge slower than $L_C^\alpha$ for $\alpha\neq0$, $L_C^\alpha$ can play the dominant role in $\kappa_{eff}$. From this point of view, $\kappa_{eff}\sim L_C^\alpha$ is not well-established, which needs further discussion on the boundary effects. Furthermore, convergent $\kappa_{eff}$ can be recovered by introducing a coexistence of fractional and standard collisions, namely,

$$\partial_t f_P+\mathbf{v}_g\cdot\nabla f_P=\theta\tau^\alpha D_t^\alpha\left(\frac{f_0-f_P}{\tau}\right)+(1-\theta)\left(\frac{f_0-f_P}{\tau}\right),\tag{25}$$

where $0\leqslant\theta<1$. This linearized BTE leads to the following constitutive equation

$$(1-\theta)\mathbf{q}+\theta\tau^\alpha D_t^\alpha\mathbf{q}+\tau\partial_t\mathbf{q}=-\kappa\nabla T\tag{26}$$

which predicts $\kappa_{eff}=\kappa/(1-\theta)$ as $t\gg\tau$.

We now consider thermodynamics in phonon heat transport, which involves entropic concepts including the entropy density $s=s(\mathbf{x},t)$, entropy flux $\mathbf{J}=\mathbf{J}(\mathbf{x},t)$, and entropy production rate $\sigma=\sigma(\mathbf{x},t)$. In the near-equilibrium region, the entropy density and entropy flux can be expressed in terms of classical irreversible thermodynamics (CIT) [40], namely,

$$s = \int c\frac{dT}{T}, \mathbf{J} = \frac{\mathbf{q}}{T}. \tag{27}$$

The entropy production rate can be calculated from the following entropy balance equation

$$\sigma = \partial_t s + \nabla \cdot \mathbf{J} = \mathbf{q} \cdot \nabla\left(\frac{1}{T}\right). \tag{28}$$

For time-independent $T$, we have $\mathbf{q} \sim t^\alpha$. It means that a stationary thermodynamic state is paired with time-dependent $\mathbf{J}$ and $\sigma$ divergent to infinity. These entropic behaviors are very different from the usual understanding in CIT that the entropy production rate commonly converges for steady-state. Thus, the validity of the CIT formulism is debatable, and the entropic functionals requires further studies. In Boltzmann-Gibbs (BG) statistical mechanics, the entropy density of phonons is given by

$$s = k_B \int [(f_P+1)\ln(f_P+1) - f_P \ln f_P]d\mathbf{k}. \tag{29}$$

One can expand Eq. (29) as

$$s = s_0 + s_1 + k_B \int o(f_P - f_0)d\mathbf{k}, \tag{30}$$

wherein

$$s_0 = k_B \int [(f_0+1)\ln(f_0+1) - f_0 \ln f_0]d\mathbf{k} \tag{31a}$$

$$s_1 = k_B \int (f_P - f_0)\ln\frac{f_0+1}{f_0}d\mathbf{k} \tag{32b}$$

Noting that $\int \hbar\omega f_P d\mathbf{k} = \int \hbar\omega f_0 d\mathbf{k}$ and $\ln\frac{f_0+1}{f_0} = \frac{\hbar\omega}{k_B T}$, we acquire

$$\partial_T s_0 = k_B \int \ln\frac{f_0+1}{f_0}\partial_T f_0 d\mathbf{k} = \frac{1}{T}\int \hbar\omega\partial_T f_0 d\mathbf{k} = \frac{c}{T}, \tag{33a}$$

$$s_1 = k_B \int (f_P - f_0)\ln\frac{f_0+1}{f_0}d\mathbf{k} = \frac{1}{T}\int \hbar\omega(f_P - f_0)d\mathbf{k} = 0 \tag{33b}$$

Then, we arrive at $s = s_0 + k_B \int O(f_P - f_0)^2 d\mathbf{k}$. It agrees with the CIT entropy density only if $k_B \int O(f_P - f_0)^2 d\mathbf{k}$ is neglected, which needs $|f_P - f_0| \ll f_0$. However, the divergence $\mathbf{q} \sim t^\alpha$ implies

$$\mathbf{q} = \int \mathbf{v}_g f_P \hbar\omega d\mathbf{k} = \int \mathbf{v}_g (f_P - f_0)\hbar\omega d\mathbf{k} \sim t^\alpha, \tag{34}$$

which means divergent $(f_P - f_0)$ as $t \to +\infty$. Therefore, $|f_P - f_0| \ll f_0$ is possibly invalid and $k_B \int O(f_P - f_0)^2 d\mathbf{k}$ cannot be neglected. It exhibits possible invalid of the CIT entropy density. The temporal derivative of Eq. (29) reads

$$\partial_t s = k_B \int \partial_t f_P[\ln(f_P+1) - \ln f_P]d\mathbf{k} \tag{35}$$

Substituting Eq. (23) into Eq. (35) yields

$$\begin{aligned}\partial_t s = &-\nabla \cdot \left\{\int \mathbf{v}_g k_B[(f_P+1)\ln(f_P+1) - f_P \ln f_P]d\mathbf{k}\right\} \\ &+ k_B \int \ln\frac{f_P+1}{f_P}\tau^\alpha D_t^\alpha\left(\frac{f_0 - f_P}{\tau}\right)d\mathbf{k},\end{aligned} \tag{36}$$

which indicates

$$\mathbf{J} = \int \mathbf{v}_g k_B[(f_P+1)\ln(f_P+1) - f_P \ln f_P]d\mathbf{k}, \tag{37a}$$

$$\sigma = k_B \int \ln\frac{f_P+1}{f_P}\tau^\alpha D_t^\alpha\left(\frac{f_0 - f_P}{\tau}\right)d\mathbf{k}. \tag{37b}$$

Similarly, Eq. (37a) can be expanded as

$$\mathbf{J} = \mathbf{J}|_{f_P=f_0} + \int \mathbf{v}_g k_B \ln\frac{f_0+1}{f_0}(f_P - f_0)d\mathbf{k} + \int \mathbf{v}_g k_B o(f_P - f_0)d\mathbf{k}. \tag{38}$$

Noting that $\mathbf{J}|_{f_P=f_0} = \mathbf{0}$ and $\int \mathbf{v}_g \hbar\omega f_0 d\mathbf{k} = \mathbf{0}$, we deduce

$$\begin{aligned}\mathbf{J} &= \frac{1}{T}\int \mathbf{v}_g \hbar\omega(f_P - f_0)d\mathbf{k} + \int \mathbf{v}_g k_B O(f_P - f_0)^2 d\mathbf{k} \\ &= \frac{\mathbf{q}}{T} + \int \mathbf{v}_g k_B O(f_P - f_0)^2 d\mathbf{k}.\end{aligned} \tag{39}$$

From Eq. (39), we fin that similar invalidity caused by $O(f_P - f_0)^2$ also exists for the entropy flux. The expansion of Eq. (37b) reads

$$\sigma = \sigma_0 + \sigma_1 + k_B \int O(f_P - f_0)^2 \tau^\alpha D_t^\alpha\left(\frac{f_0 - f_P}{\tau}\right)d\mathbf{k}, \tag{40}$$

wherein

$$\sigma_0 = k_B \int \ln\frac{f_0+1}{f_0}\tau^\alpha D_t^\alpha\left(\frac{f_0 - f_P}{\tau}\right)d\mathbf{k}, \tag{41a}$$

$$\begin{aligned}\sigma_1 &= k_B \int \frac{d}{df_0}\left(\ln\frac{f_0+1}{f_0}\right)(f_P - f_0)\tau^\alpha D_t^\alpha\left(\frac{f_0 - f_P}{\tau}\right)d\mathbf{k} \\ &= k_B \int \frac{d}{df_0}\left(\ln\frac{f_0+1}{f_0}\right)(f_P - f_0)(\partial_t f_P + \mathbf{v}_g \cdot \nabla f_P)d\mathbf{k},\end{aligned} \tag{41b}$$

The zero-order term $\sigma_0$ equals to zero:

$$\begin{aligned}\sigma_0 &= k_B \int \frac{\hbar\omega}{k_B T}\tau^\alpha D_t^\alpha\left(\frac{f_0 - f_P}{\tau}\right)d\mathbf{k} \\ &= \frac{1}{T}\tau^\alpha D_t^\alpha \int \hbar\omega\left(\frac{f_0 - f_P}{\tau}\right)d\mathbf{k} = 0.\end{aligned} \tag{42a}$$

The first-order term $\sigma_1$ can be divided into two parts: $\sigma_1 = \sigma_{10} + \sigma_{11}$ with

$$\begin{aligned}\sigma_{10} &= k_B \int \frac{\partial}{\partial f_0}\left(\ln\frac{f_0+1}{f_0}\right)(f_P - f_0)(\partial_t f_0 + \mathbf{v}_g \cdot \nabla f_0)d\mathbf{k} \\ &= \partial_t\left(\tfrac{1}{T}\right)\int \hbar\omega(f_P - f_0)d\mathbf{k} + \nabla\left(\tfrac{1}{T}\right) \cdot \int \hbar\omega\mathbf{v}_g(f_P - f_0)d\mathbf{k} \\ &= \mathbf{q} \cdot \nabla\left(\tfrac{1}{T}\right),\end{aligned} \tag{42b}$$

$$\sigma_{11} = \frac{k_B}{2}\int \frac{d}{df_0}\left(\ln\frac{f_0+1}{f_0}\right)\left[\partial_t(f_P - f_0)^2 + \mathbf{v}_g \cdot \nabla(f_P - f_0)^2\right]d\mathbf{k} \tag{42c}$$

Different from the above cases, possible invalidity of CIT arises from not only $O(f_P - f_0)^2$ but also its derivatives in $\sigma_{11}$. In summary, Eq. (23) will lead to anomalous entropic behaviors in phonon heat transport.

## 3. Comparisons with other fractional models

Upon multiplying Eq. (10) by $|\mathbf{v} - \mathbf{c}|^2$ (or $\hbar\omega$) and integrating it, we can recover Eq. (18). Combining Eq. (18) with Eq. (13) yields

$$\tau^\alpha D_t^{\alpha+1}T + \tau\partial_t^2 T = D\nabla^2 T + \frac{T|_{t=0}\tau^\alpha}{t^{\alpha+1}\Gamma(-\alpha)}, \tag{43}$$

which reduces to the Cattaneo's model as $\alpha = 0$. When the initial value term $\frac{T|_{t=0}\tau^\alpha}{t^{\alpha+1}\Gamma(-\alpha)}$ is neglected, Eq. (43) will be included by the GCE class [36]. In this class, Eq. (43) is the so-called GCE II. The difference is that the GCE II arises from the following constitutive equation rather than Eq. (13),

$$\mathbf{q} + \tau^{1-\alpha}D_t^{1-\alpha}\mathbf{q} = -\tau^{-\alpha}D_t^{-\alpha}(\kappa\nabla T). \tag{44}$$

In the GCE class, all of the initial value terms are neglected. Thereby, $D_t^m D_t^n$ equals to $D_t^{m+n}$, and Eq. (44) can recover Eq. (13) through multiplying the operator $\tau^\alpha D_t^\alpha$. Besides Eq. (44), the GCE class also introduces other fractional-order constitutive equations as follows:

$$\mathbf{q}+\tau^{1-\alpha}D_t^{1-\alpha}\mathbf{q}=-\kappa\nabla T, \tag{45a}$$

$$\mathbf{q}+\tau^{1-\alpha}D_t^{1-\alpha}\mathbf{q}=-\tau^{\alpha}D_t^{\alpha}(\kappa\nabla T), \tag{45b}$$

$$\mathbf{q}+\tau\partial_t\mathbf{q}=-\tau^{\alpha}D_t^{\alpha}(\kappa\nabla T). \tag{45c}$$

In phonon heat transport, the above constitutive equations can be derived from the following linearized BTEs, respectively,

$$\tau^{-\alpha}D_t^{1-\alpha}f_P+\mathbf{v}_g\cdot\nabla f_P=\frac{f_0-f_P}{\tau}, \tag{46a}$$

$$\tau^{-\alpha}D_t^{1-\alpha}f_P+\tau^{\alpha}D_t^{\alpha}(\mathbf{v}_g\cdot\nabla f_P)=\frac{f_0-f_P}{\tau}, \tag{46b}$$

$$\partial_t f_P+\tau^{\alpha}D_t^{\alpha}(\mathbf{v}_g\cdot\nabla f_P)=\frac{f_0-f_P}{\tau}. \tag{46c}$$

Different from Eq. (23), the above BTEs posses the standard collision term, which does not contain any memory kernel. For time-independent $T$, Eqs. (45a–c) lead to the following $\mathbf{q}$, respectively,

$$\mathbf{q}=\left[\left(t^{-\alpha}D_t^{-\alpha}\mathbf{q}\right)\big|_{t=0}\right]E_{1-\alpha,1-\alpha}\left[-\left(\frac{t}{\tau}\right)^{1-\alpha}\right]-\kappa\left(\frac{t}{\tau}\right)^{1-\alpha}E_{1-\alpha,2-\alpha}\left[-\left(\frac{t}{\tau}\right)^{1-\alpha}\right]\nabla T, \tag{47a}$$

$$\mathbf{q}=\left[\left(t^{-\alpha}D_t^{-\alpha}\mathbf{q}\right)\big|_{t=0}\right]E_{1-\alpha,1-\alpha}\left[-\left(\frac{t}{\tau}\right)^{1-\alpha}\right]-\kappa\left(\frac{t}{\tau}\right)^{1-2\alpha}E_{1-\alpha,2-2\alpha}\left[-\left(\frac{t}{\tau}\right)^{1-\alpha}\right]\nabla T, \tag{47b}$$

$$\mathbf{q}=(\mathbf{q}|_{t=0})\exp\left(-\frac{t}{\tau}\right)-\kappa\left(\frac{t}{\tau}\right)^{1-\alpha}E_{1,2-\alpha}\left(-\frac{t}{\tau}\right)\nabla T. \tag{47c}$$

In contrast with Eq. (14), Eqs. (47a–c) will converge as $t\to+\infty$, which implies convergent $\kappa_{eff}$. In the long-time limit, Eq. (47a) implies $\kappa_{eff}\to\kappa$, while for Eqs. (47b) and (47c), $\kappa_{eff}\to 0$. According to the conclusions by Compte and Metzler [36], the GCE class corresponds to superdiffusive long-time asymptotics with $\gamma>1$. Obviously, Eqs. (47–47c) can never fulfill $\beta=\gamma-1$. In usual understandings of anomalous heat conduction, superdiffusion should correspond to divergent $\kappa_{eff}$ and in the ballistic limit ($\gamma\to 2$), $\kappa_{eff}\sim L_C$. Consequently, the standard collision term, which even predicts convergence to zero for $\gamma>1$, is not applicable in superdiffusive and ballistic heat conduction. One possible reason is that Eqs. (45–45c) cannot recover the standard continuity equation. Through multiplying Eqs. (45–45c) by $\hbar\omega$ and integrating them over the wave vector space, we arrive at fractional-order continuity equations as follows

$$\tau^{-\alpha}D_t^{1-\alpha}e=-\nabla\cdot\mathbf{q}, \tag{48a}$$

$$\tau^{-\alpha}D_t^{1-\alpha}e=-\tau^{\alpha}D_t^{\alpha}(\nabla\cdot\mathbf{q}), \tag{48b}$$

$$\partial_t e=-\tau^{\alpha}D_t^{\alpha}(\nabla\cdot\mathbf{q}). \tag{48c}$$

All of the above continuity equations deviate from the standard form, yet in the linear response theory, the standard continuity equation is a fundamental assumption to derive $\beta=\gamma-1$. Power-law divergence indicates sharp enhancement of heat transport with increasing $L_C$, and surprisingly high $\kappa_{eff}$ at the macroscale, which is intriguing in thermal management. Accordingly, the fractional-order collision term is unedifying. One unsatisfactory feature of the present work is that it cannot predict the logarithmic divergence, which is universal in 2D systems. The dimensionality-dependence is not reflected either, which may be associated with $\alpha$. Moreover, the fractional-order derivative is defined by the RL operator, which contains singular memory kernel. One can replace the RL operator by other generalized forms [41–43] to overcome singularity.

## 4. Conclusions

1. The fractional-order BGK model proposed by Goychuk is discussed in superdiffusive and ballistic heat conduction, which is characterized by $\langle\Delta\mathbf{x}^2\rangle\sim t^{\gamma}$ with $1<\gamma\leqslant 2$. It is shown that the fractional-order BTE predicts a fractional-order constitutive equation in both kinetic and phonon heat transport. This constitutive equation yields divergent effective thermal conductivity, whose long-time asymptotics obeys $\kappa_{eff}\sim t^{\alpha+1}$. The scaling relation between the MSD and divergence is obtained as $\beta=\gamma-1$, which coincides with previous investigations. These coincidences exhibit the robustness of the fractional-order BTE.
2. Besides divergent effective thermal conductivity, the fractional-order BTE also indicates anomalous behaviors of the entropic functionals including entropy, entropy flux and entropy production rate. The entropy flux and entropy production rate diverge to infinity in a stationary temperature field. Through series expansions in BG statistical mechanics, we show that the entropic functionals for the fractional-order BGK model cannot be expressed by the CIT formulism. The entropic anomaly arises from the second-order term $(f_P-f_0)^2$ and its derivatives.
3. Goychuk's model leads to a governing equation belonging to the GCE class. Other constitutive equations in the GCE class can emerge from different fractional-order BTEs as well. The difference is that their collision terms remain the standard form. In contrast with Goychuk's model, these linearized BTEs lead to convergent effective thermal conductivity in superdiffusive heat conduction, and some models even predict convergence to zero. These features deviate from usual understandings on anomalous heat conduction, and thus, other models in the GCE class are not applicable.

## Conflict of interest

The authors declare no conflict of interest.

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