Guyer-Krumhansl–type heat conduction at room temperature

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Abstract – Results of heat pulse experiments in various artificial and natural materials are reported in this paper. The experiments are performed at room temperature with macroscopic samples. It is shown that temperature evolution does not follow Fourier’s law but is well explained by the Guyer-Krumhansl equation. The observations confirm the ability of non-equilibrium thermodynamics to formulate universal constitutive relations for thermomechanical processes.

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Introduction. – Since the discovery of second sound in low-temperature solids in the 1970s [1–4], there were several predictions of similar phenomena with various assumptions and conditions [5,6], including heterogeneous materials at room temperature. The first promising positive experimental results in granular and biological media were reported about Maxwell-Cattaneo-Vernotte–type heat conduction in [7,8]. However, these measurements were not confirmed, could not be repeated, the related experimental reports are contradictory [9–12]. In our interpretation this may be due to the incomplete theoretical framework.

Theory and experiment are intervening in instructive prediction and confirmation patterns in the case of non-Fourier heat conduction [13]. The first experimental discovery of the second sound in fluid helium by Peshkov [14] was theoretically predicted and motivated by the two-fluid model of Tisza and Landau [15,16]. They had introduced inertial effects for heat by assuming additional thermodynamic degrees of freedom via the two-component material model. Later, the arguments of Cattaneo and Vernotte [17,18], or previously the famous calculation of Maxwell [19], promoted a different physical background with a single-component material: the inertia of heat may have various origins.

The next important experimental step, the detection of second sound and ballistic propagation in solids, used a kinetic theory background understanding and predicted the phenomena by detailed microscopic modelling of phonons. There, the two different relaxation mechanisms in the Callaway collision integral resulted in the development of the Maxwell-Cattaneo-Vernotte–type (MCV) heat conduction by Guyer and Krumhansl [20,21]. From the point of view of experimental discovery it was a crucial step, because it provided a method to reduce the additional dissipative effects via the so-called window condition [22,23]. Here, the theoretical background, that is phonon hydrodynamics with non-Fourier effects, is a theory of rarefied gases. In the case of solids, it requires microscopic homogeneous samples and low temperature. Recently the Guyer-Krumhansl equation is intensively studied and new exact solutions and numerical solution methods were developed for various related Cauchy problems [24–29], several other theoretical researches are improving looking for observable phenomena in nanomaterials using the aforementioned background [30–32], the kinetic theory background is also further developed and refined [33,34].

The Guyer-Krumhansl (GK) equation is as follows:
\[
\tau q + q + \lambda_F \nabla T - \beta' \Delta q - \beta'' \nabla \cdot \nabla q = 0.
\] (1)
Here \(q\) is the heat flux, the current density of the internal energy in a rest frame of the continuum, \(\lambda_F\) is the Fourier heat conduction coefficient, \(\tau\) is a relaxation time, while...
\( \beta' \) and \( \beta'' \) are Guyer-Krumhansl coefficients in isotropic media. These are related to the relaxation times of the Callaway collision integral in rarefied gases. The overdot denotes the time derivative, \( \nabla \) is the space derivative, and the coefficients are considered as constant. Here, the first three terms form the Maxwell-Cattaneo-Vernotte (MCV) equation, the second and the third ones are Fourier’s law.

The conceptual foundation of looking for non-Fourier phenomena in macroscopic samples at room temperature is originated in the phenomenological theories: phonons mean free paths and relaxation times are negligible in sand or frozen meat in heat conduction at room temperature. Therefore, among the many phenomenological ideas leading to and explaining the origin of the MCV equation (see, e.g., [5,6,35–39]), non-equilibrium thermodynamics plays a distinguished role. There (1) is a so-called constitutive equation, that expresses the material properties and restricted by the second law. At the same time, it is a partial differential equation for the time and space evolution of the heat flux. The validity of a constitutive equation derived from the framework of non-equilibrium thermodynamics is based on the conditions used in the derivation. Therefore, classical equations, like Fourier’s law, are valid as long as the basic continuum balances, the local equilibrium and the second law are valid [40,41]. This is universality in the sense, that the constitutive equations are independent of the microscopic details. For MCV-type heat conduction, the local equilibrium is violated and the respective non-equilibrium thermodynamic theory, called Extended Thermodynamics, is well known [23,42]. Due to the pure phenomenological derivation [43], the experimental strategy aiming to discover non-Fourier effects in heterogeneous macroscopic samples at room temperature was seemingly justified. However, the above-mentioned experiments did not lead to reproducible results, second sound was not observed unambiguously in macroscopic samples at room temperature.

In the framework of Extended Thermodynamics, the Guyer-Krumhansl equation can be obtained by a modification of the entropy flux in all approaches, usually together with other assumptions [44,45]. A minimal functional deviation of the entropy flux from the local equilibrium — introducing Nyiri multipliers [46] — leads to the GK equation without any further ado, and it is also straightforward to obtain most of the viable suggested generalisations of Fourier’s law and MCV equation in a uniform framework [47], which is also compatible with kinetic theory [48].

Motivated by the universality of non-equilibrium thermodynamics, most recently an experimental-theoretical study has been performed in order to identify suitable qualitative signatures of detecting non-Fourier heat conduction beyond the MCV equation in heterogeneous, macroscopic samples at room temperature [49–54]. The first indication of the effect was measured on artificial samples, with alternating layers of good and bad heat conductors parallel to the heat flux [55]. However, there an adiabatic boundary was assumed and the effect is less significant, when considering the cooling of the sample. Here, in this paper, we report experimental results of heat pulse measurements on various artificial and natural heterogeneous materials, where the GK-type heat conduction is inevitable also with considering the cooling of the sample.

In the first part, the experimental setup is reviewed and the basic equations are shown. Then a non-dimensional form is introduced with heat pulse boundary conditions and the results of the measurements are presented and discussed. Subsequently simulation results are shown, first demonstrating that the GK equation reproduces the experimental data. Then, a qualitative explanation of the GK-type heat conduction is shown by a detailed simulation of the regular heterogeneity of the simplest sample.

**Heat pulse experiment and Guyer-Krumhansl model.** – In our simple experimental device, a flash lamp generates the heat pulse at the front end of the sample, and the temperature is measured by a pin-thermocouple (K-type) at the rear end. The thermocouple and the detector part are insulated from the heat pulse and from the electromagnetic noises. The heat pulse is measured directly at the front end by a photovoltaic cell, triggering data acquisition. A typical pulse shape is triangular and is 10 ms long.

A sketch of the experimental setup is shown in fig. 1. The thickness of the studied specimens is smaller than their diameter and the front face is homogeneously heated by the heat pulse, therefore, heat conduction can be considered one-dimensional.

At the front side a thin black coating is applied to ensure uniform boundary conditions, as well as to eliminate the transparency of the sample. At the rear side silver painting is used, therefore, the thermocouple measures an effective temperature. The effect of the coating is negligible according to control measurements. The experimental
device was calibrated by using several samples with known heat diffusivity.

Apparently, in the above experiment, the heat conduction is one-dimensional. Therefore, the balance of internal energy for rigid heat conductors can be written as

$$\rho c \frac{\partial T}{\partial t} + \frac{\partial q}{\partial x} = 0.$$  \hspace{1cm} (2)

Here $c$ is the specific heat, $T$ is the temperature, and the specific internal energy is $e_{sp} = cT$. $\rho$ is the density, and $q$ is the heat flux in the direction of the heat propagation. The evolution equation of the heat flux is the one-dimensional version of (1):

$$\tau \frac{\partial q}{\partial t} + q + \lambda_F \frac{\partial T}{\partial x} - l^2 \frac{\partial^2 q}{\partial x^2} = 0.$$  \hspace{1cm} (3)

Here $k$ is the Fourier heat conduction coefficient, $\tau$ is the relaxation time and $l^2 = \beta' + \beta''$ is a non-negative material parameter of the GK equation, and is expressed with the help of a characteristic length scale $l$.

Remarkable and instructive is to recognise a hierarchical structure in the above system of equations, (2), (3), when expressed for the temperature [52,56,57]. It is best seen by eliminating the heat flux and rearranging the system as follows:

$$\tau \frac{\partial T}{\partial t} \left( \frac{\partial T}{\partial t} - b \alpha \frac{\partial^2 T}{\partial x^2} \right) + \left( \frac{\partial T}{\partial t} - \alpha \frac{\partial^2 T}{\partial x^2} \right) = 0,$$  \hspace{1cm} (4)

where $\alpha = \frac{k}{\rho c}$ is the thermal diffusivity and $b = \frac{l^2}{\tau^2}$ is the coefficient characterising the deviation from Fourier heat conduction. One can observe that solutions of the Fourier equation are solutions of (4), whenever $b = 1$, that is

$$\alpha = \frac{l^2}{\tau}.$$  \hspace{1cm} (5)

Hereafter we will call this formula the Fourier resonance condition. If $b < 1$ then the solutions of (2), (3) show wavelike characteristics; if $b > 1$ then the solutions are over-diffusive [48,58].

We are looking for solutions of (2), (3) with heat pulse boundary condition and surface heat exchange at the front side. Therefore,

$$q(x, 0, t) = \begin{cases} q_{\text{max}} \left(1 - \cos \left(2\pi \frac{t}{t_p}\right)\right) - B(T(0, t) - T_0), & \text{if } 0 < t \leq t_p, \\ -B(T(0, t) - T_0), & \text{if } t > t_p. \end{cases}$$

Here $t_p$ is the pulse duration, $T_0$ is the ambient temperature and $B$ is the heat exchange coefficient. The particular shape of the pulse does not influence the effect as long as the duration of the pulse is much shorter than the characteristic time scale of the experiment [59]. Therefore here we can use the smooth cosine function for the sake of stability of numerical solution. The backside boundary is considered adiabatic: $q(L, t) = 0$, because of the insulating cover. Initially, the temperature distribution is uniform and the heat flux is zero along the sample, that is, $T(x, 0) = T_0$ and $q(x, 0) = 0$, this is provided by the measurement protocol, with 30–60 minutes temperature equilibration periods between the measurements.

Suitable dimensionless forms of the variables are:

$$\dot{i} = \frac{t}{t_p}, \quad \dot{x} = \frac{x}{L};$$

$$\dot{q} = \frac{q}{q_{\text{max}}}, \quad \text{where } q_{\text{max}} = \frac{1}{t_p} \int_0^{t_p} q_0(t) \, dt;$$

$$\dot{T} = s \frac{T - T_0}{T_{\text{end}} - T_0}, \quad \text{where } T_{\text{end}} = T_0 + q_{\text{max}} t_p, L \rho c,$$

and $$s = \frac{T_{\text{ref}} - T_0}{T_{\text{end}} - T_0}.$$  \hspace{1cm} (6)

where $q_0(t) = q(x = 0, t) + B(T(0, t) - T_0)$ is the boundary condition without cooling, $T_{\text{ref}}$ is a reference temperature, e.g., the measured maximum temperature, which is different from the limiting adiabatic temperature, $T_{\text{end}}$, because of the cooling. The dimensionless parameters are, consequently,

$$\dot{\tau} = \frac{\tau}{t_p}; \quad \dot{\alpha} = \alpha \frac{t_p}{L^2}; \quad \dot{\beta} = \frac{\dot{l}^2}{\tau \dot{\alpha}} = b; \quad \dot{B} = B \frac{t_p}{\rho c L}.$$  \hspace{1cm} (7)

Accordingly, the non-dimensional form of the equations is

$$s \frac{\hat{T}}{\partial t} + \hat{q} = 0,$$  \hspace{1cm} (8)

$$\dot{\alpha} \hat{T} + \hat{q} + s \hat{\alpha} \frac{\partial \hat{T}}{\partial \hat{x}} - b \dot{\alpha} \frac{\partial^2 \hat{q}}{\partial \hat{x}^2} = 0.$$  \hspace{1cm} (9)

The boundary and initial conditions are:

$$\hat{q}(x = 0, t) = \begin{cases} 1 - \cos \left(2\pi \frac{t}{t_p}\right) - s \hat{B} \hat{T}(0, t), & \text{if } 0 < \dot{i} \leq 1, \\ -s \hat{B} \hat{T}(0, t), & \text{if } \dot{i} > 1. \end{cases}$$

Here the parameters $\tau$, $\alpha$ and $B$ characterise the material, the temperature scale, $s$, and the heat exchange coefficient, $B$, do not.

**Measurements.** – Among the investigated samples four had been chosen, where the deviation from Fourier heat conduction was the most apparent. These were a capactor, the same as in [55], a crystalline limestone sample from Villány, southern Hungary, a leucocratic rock sample and a metal foam sample (see fig. 2). The corresponding backside temperature is shown in figs. 3 and 4. The system of equations (8), (9) is solved and the parameters are fitted to the data with the built-in nonlinear regression algorithm of Mathematica 11.0, using the solution of the system of partial differential equations as an input function. On the figures the non-dimensional temperature is scaled approximately to the adiabatic limiting temperature ($0.9 T_{\text{max}}$), the data is slightly smoothed with 3-point
Fig. 2: (Color online) The heterogeneous structure of the measured materials. From left to right: capacitor, limestone from Villány, metal foam, leucocratic rock with slires. The front sides of the samples are shown before the black coating is applied.

Fig. 3: (Color online) Top: an artificial, layered capacitor sample; bottom: limestone from Villány (southern Hungary). The experimentally measured data of the backside temperature as the function of time. The solid line is the best-fitted solution of the Guyer-Krumhansl equation, the dashed line is the best-fitted Fourier equation with correct asymptotics.

Table 1: Dimensionless parameters.

| Sample            | $\hat{\alpha} \times 10^3$ | $\hat{\tau}$ | $b$  |
|-------------------|-----------------------------|--------------|------|
| Capacitor         | 1.40                        | 95.4         | 2.23 |
| Limestone         | 1.124                       | 99.1         | 2.17 |
| Metal foam 1      | 0.912                       | 40.2         | 3.04 |
| Leucocratic       | 1.56                        | 132.0        | 1.77 |

Table 2: Physical parameters.

Running average. The solid lines show the best GK fit and the dashed line is the best Fourier fit with the correct asymptotics.

The dimensionless parameters are given in table 1 and the related physical parameters in table 2.

All measurements indicate an overdiffusive, $b > 1$, Guyer-Krumhansl-type heat conduction.

Fig. 4: (Color online) Top: metal foam sample 1; bottom: leucocratic rock with slires. The experimentally measured data of the backside temperature as the function of time. The solid line is the best-fitted solution of the Guyer-Krumhansl equation, the dashed line is the best-fitted Fourier equation with correct asymptotics.

Fig. 5: (Color online) Geometry of the sample. The red color is Al, the blue is polystyrol.

**Simulation of heterogeneity.** In case of the capacitor sample the effect of regular heterogeneity can be calculated directly. The geometry of the sample is shown in fig. 5. It is assumed, that the cylindrical symmetry is not important and a simpler arrangement can show the related effect. Also an aluminium layer is applied at the upper (rear) side instead of the silver coating. This geometry is expected to reproduce the measured backside temperature of the heat pulse experiment with the capacitor sample. The capacitor is not symmetric, the aluminium and polystyrol layers are rolled and continuous, therefore, a two-dimensional representation seems to be proper approximation. The thicknesses of the layers are $5\mu m$ and $15\mu m$ for the aluminium and for the polystyrol, respectively.

The applied material parameters are given in table 3.

The Fourier equation ((4) with $\tau = 0$) is solved numerically with a finite volume algorithm dealing excellently with sharp interfaces and material heterogeneity [60–63]. The time step was chosen $\Delta t = 0.01 t_p = 10^{-4} s$ and the spatial dimensions were $185 \times 40$ space steps, 5 and 15 steps for the aluminium and polystyrol, respectively. The calculations are performed for $10 s = 100000$ time steps in two cases. In the first case a pure aluminium sample was assumed and the comparison with the data of Both et al. [55] is shown on the left side of fig. 6. The measured and...
Table 2: Dimensional parameters. $\alpha_F$ is the thermal diffusivity from Fourier equation (dashed line), $\alpha_{\text{GK}}$ is the thermal diffusivity with the Guyer-Krumhansl equation (solid line).

| Sample  | $L$ (mm) | $\alpha_F \times 10^6$ ($\text{m}^2/\text{s}$) | $\alpha_{\text{GK}} \times 10^6$ ($\text{m}^2/\text{s}$) | $\tau$ (s) | $l$ (mm) |
|---------|---------|---------------------------------|---------------------------------|-----------|---------|
| Capacitor | 3.9     | 3.45                            | 2.13                            | 0.954     | 2.79    |
| Limestone| 1.7     | 0.45                            | 2.950                           | 0.991     | 0.80    |
| Metal foam 1 | 5.1     | 3.04                            | 2.373                           | 0.402     | 1.70    |
| Leucocratic | 1.75    | 7.14                            | 4.77                            | 1.32      | 1.06    |

Table 3: Material parameters.

|                | Density (kg/m$^3$) | Heat capacity (J/kgK) | Conductivity (W/mK) | Diffusivity ($\text{m}^2/\text{s}$) |
|----------------|---------------------|------------------------|----------------------|--------------------------------------|
| Aluminium      | 2707                | 905                    | 237                  | $9.61 \times 10^{-5}$                |
| Polystyrol     | 1040                | 1350                   | 0.15                 | $10^{-7}$                            |

Fig. 6: (Color online) Microstructure simulation for the capacitor sample. Top: pure aluminium; bottom: heterogeneous aluminium-polystyrol two-dimensional simulation compared to the measured data.

Simulated dimensionless backside temperature is shown as a function of time by the thin and thick solid lines, respectively. With the geometry of fig. 5, the material parameters of table 3, the backside temperature is shown on the right side of fig. 6.

One can see that for pure aluminium, after an initial coincidence, the slope of the computed curve becomes very different from the experimental values. For the heterogeneous case the simulated curve is closer to the experimental one, but the difference still exists. In computations, the temperature at the upper boundary is slowly decreased due to the huge difference in conductivities of aluminium and polystyrol in spite of the adiabatic boundary conditions. However, the difference in values of temperature is very small and cannot be registered in experiments. It is illustrated by fig. 7, where the temperature field corresponds to 100000 time steps.

Summary and discussion. – We have presented backside temperature data of heat pulse experiments at room temperature in several artificial and natural macroscopic, heterogeneous specimen with various internal structure at room temperature. The presented data indicates non-Fourier heat conduction in the overdiffusive, $b > 1$, regime of the GK equation, where wavelike phenomena are utterly suppressed. The parameters of the simulated solution indicate large differences in the measured Fourier heat conduction coefficients with pure Fourier model fitting or with a GK model fitting (see table 2). The cooling of the sample is considered in the calculations. The natural length parameter of the fitted GK equation is in the order of magnitude of the size of the sample and not
at the length scale of the observed heterogeneity (width of the layers, pore size or hole size).

Detailed two-dimensional simulation of the capacitor sample considering the regular heterogeneity with Fourier equation could not reproduce the experimental observations. This deviation indicates that the pure phenomenological GK fit with effective parameters can consider further, hidden heterogeneity in the material (e.g., the distributed thermal resistance between the aluminium and polystyrol layers).

The observed deviation from Fourier’s law at room temperature is modelled by the same theory, by the GK equation, as at low temperatures in homogeneous samples. The mechanisms are evidently different, the apparent common aspect is probably the presence of various heat conduction channels. The two relaxation terms in case of Callaway collision integral provides two propagation mechanisms for phonons inside spatially homogeneous samples. The aluminium and polystyrol in case of the capacitor, the aluminium and air in case of a metal foam are probably responsible for a similar effect that can be modelled by the GK equation as an effective description. In the case of natural, and seemingly homogeneous, rock samples there is no apparent simple explanation, because of the lack of evident, spatially separated components with big differences in the heat conduction coefficients. The nonequilibrium thermodynamic framework, extending classical irreversible thermodynamics with a minimal deviation from local equilibrium, explains the observed universality of the GK equation [47,48,64].

It is also worth remarking, that other phenomenological models, like the dual-phase-lag equation [65,66] or two-temperature coupled Fourier models (see, e.g., [67,68]) may be applied for modelling our experiments and this possibility is open. However, some of these models are particular realizations of the internal-variable–based thermodynamic approach, as it was demonstrated in [47]. These are for example the various versions of the two-temperature model and also the first approximation of the dual-phase-lag equation. Some other models are thermodynamically not consistent, for example the complete, delay-type dual-phase-lag equation [69].

A practical importance of our observations is the improvement of technological data of transient and stationary heat conduction in heterogeneous structures. This can simplify and guide microstructure-based detailed analyses and computations.

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