

# THEORIES AND HEAT PULSE EXPERIMENTS OF NON-FOURIER HEAT CONDUCTION

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**ABSTRACT.** The experimental basis and theoretical background of non-Fourier heat conduction is shortly reviewed from the point of view of non-equilibrium thermodynamics. The performance of different theories is compared in case of heat pulse experiments.

## 1. INTRODUCTION

There are several extensive reviews of heat conduction beyond Fourier [1, 2, 3, 4, 5, 6]. When non-equilibrium thermodynamics is concerned, usually Extended Thermodynamics is in the focus of surveys in this subject. In this short paper heat conduction phenomena is reviewed from a broader perspective of non-equilibrium thermodynamics. The performance of different theories is compared on the example of heat pulse experiments.

There are two preliminary remarks to underline the particular point of view of this work. The first remark concerns the so called heat conduction paradox of the infinite speed of signal propagation in Fourier theory. Infinite propagation speeds are frequently mentioned as a disadvantage [7, 8] and this is a main motivation for looking symmetric hyperbolic evolution equations in general [9]. However

- the relativistic formulation of heat conduction is a different matter and parabolic equations may be compatible with relativity [10, 11, 12].
- The finite characteristic speeds of hyperbolic equations are determined by material parameters. Therefore, in principle, in nonrelativistic spacetime they can be larger than the speed of light.
- In case of parabolic equations the speed of real signal propagation is finite, e.g. because the validity condition of a continuum theory determines a propagation speed. Moreover, the sensitivity of the experimental devices is finite, therefore they can measure only finite speeds [13, 10, 14, 15].

Our second remark is about the role of non-equilibrium thermodynamics as a frame theory of several physical disciplines. Here the fundamental assumption is the validity of the second law of thermodynamics in the form of an entropy balance with nonnegative production. If microscopic or mesoscopic theories do not want to violate macroscopic second law, they must suit the requirements derived from the phenomenological conditions. This is the extension of the classical universality of the absolute temperature. The validity of the second law is a weak assumption from a physical point of view but introduces remarkably strict restrictions for the evolution equations.

According to these arguments it is no unreasonable to look for parabolic or at least non-hyperbolic extensions of the Fourier equation of heat conduction that fulfill basic principles of continuum physics.

In the following section we survey the most important experimental observations of non-Fourier heat conduction. Then the relevant kinetic and thermodynamic theories are surveyed. Their assumptions, properties and modeling capabilities are compared on the example of heat pulse experiments.

## 2. EXPERIMENTS

**2.1. Helium II.** The first experimental observation of non-Fourier heat conduction was the measurement of wave like propagation of heat in liquid Helium II by Peshkov [16]. The experiment was motivated by two-fluid theories of Tisza [17] and Landau [18]. Landau suggested that wave like propagation is a property of phonon gas and introduced the terminology "second sound". The works of Cattaneo, Morse-Feshbach and Vernotte [7, 19, 20] indicated that the related phenomena may be more general, there is an "inertia of heat".

**2.2. Low temperature solids.** Phonon gas theory predicted the existence of second sound in solids. The key aspects of the observation were the modeling of phonon scattering by kinetic theory [21, 22, 23] and the application of heat pulses. The derivation of a dissipative extension of the Maxwell-Cattaneo-Vernotte equation from kinetic theory, the Guyer-Krumhansl equation [22], provided the "window condition", when dissipation is minimal. A careful preparation of crystals with particular properties resulted in observation of second sound in solid  $^3\text{He}$ ,  $^4\text{He}$ , NaF and Bi crystals [24, 25, 26, 27]. In some experiments second sound and ballistic propagation – heat pulses propagating with the speed of sound – were observed together [28]. At lower temperatures ballistic propagation appears without second sound.

The correct modeling of the observed parallel ballistic, wave like and diffusive propagation of heat in a uniform theoretical framework is the most important benchmark of the theories.

**2.3. Heterogeneous materials at room temperature.** The low temperature heat pulse measurements in dielectric crystals exploit well understood microscopic mechanisms. At room temperature various dissipative processes suppress these effects. However, some experiments with heterogeneous materials indicated the possibility of non-Fourier heat conduction at room temperature, too [29, 30]. These experiments were not confirmed, more properly the attempts of exact reproduction of these experiments are contradictory [31, 32, 33, 34].

**2.4. Small size.** Nano heat conduction is a popular research field with interesting new observations [35]. Non-Fourier effects are in principle enhanced by reducing the size of the samples and the speed of the phenomena [36, 37, 38]. Guyer-Krumhansl equation is extensively analyzed in this respect [39, 40].

**2.5. Relativistic experiments.** Relativistic fluid effects became experimentally available in quark-gluon plasma. Experiments in RHIC and LHC confirmed the existence of a dissipative relativistic fluid [41]. In relativistic theories the inertial effects cannot be neglected, viable theories incorporate wave like heat propagation, due to stability problems [42, 43, 44]. However, in case of ultrarelativistic speeds

the flow is energy dominated, therefore it is reasonable to use Landau-Lifshitz flow-frame, where propagation of energy defines the flow. This choice eliminates the possibility of heat conduction and only viscous dissipation is possible. According to thermodynamic arguments, the choice of flow-frames is not arbitrary and in case of lower speeds heat conduction may be separated from viscous dissipation [45].

### 3. THEORIES

The three main approaches are kinetic theory, non-equilibrium thermodynamics and continuum mechanics combined with non-Fourier heat conduction. Up to now the relation of these general approaches is not clear. Fortunately these general frame theories offer several possibilities of comparison, validation and extension far beyond heat conduction phenomena. In particular we will see, that the performance of these theories in modeling ballistic and diffusive heat propagation is different.

### 4. KINETIC THEORY

As it was already mentioned, kinetic theory played an important predictive role in understanding and designing proper experiments with the help of identified microscopic heat propagation mechanisms. In particular the relation of pure kinetic and continuum approaches is important in this respect. Kinetic theory introduces a hierarchical structure of macroscopic field quantities with coupled balance form evolution equations. In this balance structure current density at a given level is a source density at the next level. The tensorial order of the  $n$ -th variable is  $n$ . For phonons with the Callaway collision integral one may obtain the following system of equations in one spatial dimension ([9] p349):

$$(1) \quad \partial_t u_n + \frac{n^2}{4n^2 - 1} c \partial_x u_{n-1} + c \partial_x u_{n+1} = \begin{cases} 0 & n = 0, \\ -\frac{1}{\tau_R} u_1 & n = 1, \\ -\left(\frac{1}{\tau_R} + \frac{1}{\tau_N}\right) u_n & n \geq 2. \end{cases}$$

Here  $\partial_t$  and  $\partial_x$  are the time and space derivatives,  $u_n$  is related to the  $n$ th momentum of the one particle probability distribution function by constant multipliers,  $\tau_R$  and  $\tau_N$  are relaxation times,  $c$  is the Debye speed. A truncated hierarchy at the third moment,  $u_n = 0$  of  $n \geq 3$ , results in the following set of equations:

$$\begin{aligned} (2) \quad & \partial_t u_0 + c \partial_x u_1 = 0, \\ (3) \quad & \partial_t u_1 + \frac{1}{3} c \partial_x u_0 + c \partial_x u_2 = -\frac{1}{\tau_R} u_1, \\ (4) \quad & \partial_t u_2 + \frac{4}{15} c \partial_x u_1 = -\left(\frac{1}{\tau_R} + \frac{1}{\tau_N}\right) u_2. \end{aligned}$$

The macroscopic fields are introduced by the following definitions: the energy density  $e = \hbar c u_0$ , heat flux  $q = \hbar c^2 u_1$  and the moment of the heat flux  $Q = \hbar c^2 u_2$ . One may apply the (approximate) caloric equation of state  $e = \rho \hat{c} T$  with constant density  $\rho$  and specific heat  $\hat{c}$ . Moreover, it is convenient to redefine the coefficients by introducing the  $q$ -relaxation time  $\tau_q = \tau_R$ , the  $Q$ -relaxation time  $\tau_Q = \frac{\tau_R \tau_N}{\tau_R + \tau_N}$ , the Fourier heat conduction coefficient  $\lambda = \rho \hat{c} \tau_R c^3 / 3$  and two additional phenomenological coefficients  $k_{21} = \tau_R c^2$  and  $k_{12} = 4\tau / 15$ . Then the

previous equations may be written as:

$$(5) \quad \partial_t e + \partial_x q = 0,$$

$$(6) \quad \tau_q \partial_t q + \lambda \partial_x T + \kappa_{21} \partial_x Q = -q,$$

$$(7) \quad \tau_Q \partial_t Q + \kappa_{12} \partial_x q = -Q.$$

If  $\tau_Q = 0$ , one may obtain the Guyer-Krumhansl equation eliminating  $Q$  from (6)-(7). If  $\kappa_{21} = 0$  then (6) is the Maxwell-Cattaneo-Vernotte equation.

In Rational Extended Thermodynamics (RET) the coefficients of the above set of field equations are calculated exactly as it was shown above. Remarkable that one needs only three parameters, the Debye speed and the R and N process related relaxation times [9, 46, 6].

Extended Irreversible Thermodynamics (EIT) is a related but different approach, where the structure of the equations is important, but the values of the coefficients are considered mostly undetermined, therefore e.g. in (5)-(7) the number of parameters is higher [8, 47, 46]. In RET the microstructure is unavoidable in EIT the microstructure is flexible. In RET the coefficients are to be calculated from a microscopic model, in EIT some coefficients are to be measured. RET is a local theory by construction, EIT may be weakly nonlocal. RET is strictly compatible with kinetic theory, EIT is weakly compatible with kinetic theory. In EIT the derivation of the Guyer-Krumhansl equation either refers to the momentum hierarchy [47] or weakly nonlocal extensions in a pure non-equilibrium thermodynamic framework [48, 40].

The experimental results of heat conduction of mixed ballistic-wavy-diffusive propagation can be modeled in this framework. However, with the restriction of using only 3 parameters RET requires about 30 (!) moments in order to obtain correct propagation speeds both for ballistic heat propagation and second sound [49, 9]. In EIT, considering some coefficients as phenomenological parameters, in principle it is possible to incorporate correct propagation speeds for the second sound and also for the ballistic phonons with the above set of equations. However, in this case one cannot stop at the level of the Guyer-Krumhansl equation, and, most importantly, more than 3 parameters are required. A hierarchical phenomenological background theory is necessary to justify these assumptions.

## 5. NON-EQUILIBRIUM THERMODYNAMICS

Irreversible thermodynamics as field theory was established by Eckart, with the assumption of local equilibrium [50, 51, 52, 53]. Maxwell-Cattaneo-Vernotte type extensions require a deviation from local equilibrium. In this paper *non-equilibrium thermodynamics* is a collective nomination of thermodynamic theories based on the direct exploitation of the entropy inequality with or without the requirement of local equilibrium. Classical Irreversible Thermodynamics, Extended Thermodynamics, Thermodynamics with Internal Variables are special theories of non-equilibrium thermodynamics [46].

The first basic assumption extending non-equilibrium thermodynamics beyond local equilibrium is that the thermodynamic fluxes of Classical Irreversible Thermodynamics are state variables. Then Maxwell-Cattaneo-Vernotte equation is a consequence of the second law by thermodynamical flux-force linearization. This idea was first proposed by Müller [54]. One of Müller's argument was the compatibility with kinetic theory. An other independent way is based on the seminal

treatment of discrete systems by Machlup and Onsager, who introduced kinetic energy of thermodynamic state variables [55]. The continuum generalization of Gyarmati is based on variational considerations and therefore introduced thermodynamic fluxes as independent variables instead of time derivatives [56]. Later on this idea was further generalized by introducing arbitrary internal variables for the same purpose [57, 58].

As it was mentioned previously, thermodynamic fluxes as state variables may lead to the Maxwell-Cattaneo-Vernotte equation, but to obtain the Guyer-Krumhansl equation requires further assumptions. The modification of the entropy current is a straightforward idea in this respect. The idea, that the entropy current density is not always the heat flux divided by the temperature is natural in mixtures and also in Extended Thermodynamics [42], but may be natural in other generalizations, too [59, 60, 61]. To obtain the non-equilibrium thermodynamic counterpart of (6)-(7) it is reasonable to assume that the additional fields are the heat flux vector  $q$  and a second order tensorial internal variable  $Q$  and use the Nyíri form generalization of the entropy current density [62, 63].

In this paper we restrict ourselves to rigid heat conductors in a one dimensional treatment, therefore the time derivatives are partial and the space derivatives are one directional. For a more complete treatment see [64]. Our starting point is the balance of entropy, which results in nonnegative production when constrained by the balance of internal energy (5):

$$(8) \quad \partial_t s + \partial_x J \geq 0.$$

Here  $s$  is the entropy density and  $J$  is the entropy current density.

Deviation from local equilibrium will be characterized by two basic constitutive hypotheses:

- We assume that the entropy density depends quadratically on the additional fields:

$$(9) \quad s(e, q, Q) = s_{eq}(e) - \frac{m_1}{2} q^2 - \frac{m_2}{2} Q^2,$$

where  $m_1$  and  $m_2$  are constant, nonnegative material coefficients. The derivative of the local equilibrium part of the entropy function  $s_{eq}$  by the internal energy is the reciprocal temperature:

$$\frac{ds_{eq}}{de} = \frac{1}{T}.$$

The quadratic form is introduced for the sake of simplicity and may be considered as a first approximation. The coefficients  $m_1$  and  $m_2$  are non-negative because of the concavity of the entropy function, that is, thermodynamic stability.

- We assume that the entropy flux is zero if  $q = 0$  and  $Q = 0$ . Therefore it can be written in the following form:

$$(10) \quad J = bq + BQ.$$

Here  $b$  is originated from a second order tensorial constitutive function and  $B$  from a third order one. They are the current multipliers [62].

Here the basic fields are  $T, q$  and  $Q$ , the constitutive functions are  $b$  and  $B$ . The entropy production can be calculated accordingly:

$$\begin{aligned}
\partial_t s + \partial_x J &= -\frac{1}{T} \partial_x q - m_1 q \partial_t q - m_2 Q \partial_t Q + b \partial_x q + \\
&\quad q \partial_x b + B \partial_x Q + Q \partial_x B \\
(11) \quad &= \left(b - \frac{1}{T}\right) q + (\partial_x b - m_1 \partial_t q) q - (\partial_x B - m_2 \partial_t Q) Q + B \partial_x Q \geq 0.
\end{aligned}$$

In the last row the first and third terms are products of second order tensors, the second term is a product of vectors and the last term is of third order ones. It is not apparent in our one dimensional simple notation. The time derivatives of the state variables  $q$  and  $Q$  represent their evolution equations, which are considered as constitutive quantities together with the current multipliers  $b$  and  $B$ . Therefore, one can identify four thermodynamic forces and currents in the above expression and assume linear relationship between them in order to obtain the solution of the entropy inequality. In case of isotropic materials only the second order tensors can show cross effects, the vectorial and third order tensorial terms are independent.

The linear relations between the thermodynamic fluxes and forces result in the following constitutive equations:

$$(12) \quad m_1 \partial_t q - \partial_x b = -l_1 q,$$

$$(13) \quad m_2 \partial_t Q - \partial_x B = -k_1 Q + k_{12} \partial_x q,$$

$$(14) \quad b - \frac{1}{T} = -k_{21} Q + k_2 \partial_x q,$$

$$(15) \quad B = n \partial_x Q.$$

The  $l_1, k_1, k_2, n$  coefficients are nonnegative and also  $K = k_1 k_2 - k_{12} k_{21} \geq 0$ , because of the entropy inequality (11). The constitutive equations (12)-(15) together with the energy balance (5) and the caloric equation of state  $e = \rho \hat{c} T$  form a solvable set of equations. Moreover, in case of constant coefficients one can easily eliminate the current multipliers by substituting them from (14)-(15) into (12)-(13) and obtain:

$$(16) \quad m_1 \partial_t q + l_1 q - k_2 \partial_x^2 q = \partial_x \frac{1}{T} - k_{21} \partial_x Q,$$

$$(17) \quad m_2 \partial_t Q + k_1 Q - n \partial_x^2 Q = k_{12} \partial_x q.$$

Here  $\partial_x^i$  denotes the  $i$ -th partial derivative by  $x$ . (16)-(17) is identical to (6)-(7) of the previous section introducing  $\tau_q = m_1/l_1$ ,  $\lambda = (l_1 T^2)^{-1}$ ,  $\kappa_{21} = k_{21}/l_1$ ,  $\tau_Q = m_2/k_1$  and  $\kappa_{12} = k_{12}/k_1$ . If we assume that the coefficients  $k_2$  and  $n$  are zero we obtain exactly (6)-(7). For the correct sign of the coefficients there one cannot assume reciprocity here. Guyer-Krumhansl equation is a special case when  $n = 0$ ,  $m_2 = 0$  and Maxwell-Cattaneo-Vernotte is obtained if  $k_2 = 0$  and either  $k_{12} = 0$  or  $k_{21} = 0$  in addition. Several other heat conduction equations are obtained in this framework. Jeffreys type is included if  $n = 0$ ,  $m_1 = 0$ ,  $k_2 = 0$  and either  $k_{12} = 0$  or  $k_{21} = 0$ . A Cahn-Hilliard type equation is derived if  $n = 0$ ,  $m_1 = 0$  and  $m_2 = 0$  [65]. With suitable constitutive assumptions several nonlinear equations fit in this general framework like the thermomass transport equation [66], the nonlinear Maxwell-Cattaneo-Vernotte equation derived in [39] and also the nonlinear Guyer-Krumhansl equation derived in [67].

It is remarkable that the Green-Naghdi model III and II cannot be obtained here, however it is a valid special case if a general vectorial internal variable is introduced instead of the heat flux [68]).

## 6. RATIONAL THERMOMECHANICS AND SOME OTHER THEORIES

The common property of this group is the lack of compatibility with kinetic theory. The rational approaches are rigorous from mathematical point of view, require Noll type material frame indifference and insist to classical entropy current density as heat flux divided by temperature.

**6.1. Second viscosity.** Both ballistic propagation and second sound can be reasonably modeled with the help of second viscosity [69, 70, 71]. Second viscosity may have an imaginary part and its origin looks like essentially a kind of internal variable theory developed on the basis of thermostatics before the advent of non-equilibrium thermodynamics. The idea and the method is originally from Mandelstam and Leontovitch [72] and is well described in [73].

**6.2. Jeffreys type equation.** Jeffreys type equation was first suggested by Joseph and Preziosi [1] assuming a delayed transport in heat conduction. Later on this constitutive equation become very popular, because it improves the properties of Maxwell-Cattaneo-Vernotte equation from many point of view and because it can be obtained from simple (but sometimes unacceptable) assumptions [74, 38, 75]. For one dimensional heat pulse experiments the Jeffreys type equation is identical with the Guyer-Krumhansl equation.

**6.3. Green-Naghdi equations.** Green and Naghdi derived a particular constitutive equation for the heat flux. They have introduced an internal variable, called thermal displacement rate, with the particular interpretation being the time derivative of the temperature [76]. The corresponding equations coupled to the momentum balance are modeling well ballistic propagation and second sound [77, 78, 79, 80]. A particular case of the model of Green and Naghdi is the existence of heat conduction without dissipation.

**6.4. Semi-empirical temperature of Cimmelli and Kosínski.** The semi-empirical temperature is a scalar internal variable, in the framework of a weakly nonlocal theory [81, 82, 5]. Coupled to mechanics it reproduces ballistic-wave like-diffusive propagation of heat [83]. However, in this theory the constitutive theory (both the evolution of internal variable and the heat flux – temperature relation) is postulated directly.

**6.5. Rational thermomechanics.** There are many attempts to incorporate Maxwell-Cattaneo-Vernotte type heat conduction in the framework of rational thermomechanics [1, 2, 4]. It is remarkable that a nonlinear version of Maxwell-Cattaneo-Vernotte equation seems to fail the test of proper modeling the experiment, because of the lack of a certain type of dissipation, characteristic in Guyer-Krumhansl or Jeffreys type models [84, 49]. The theory could not meet the challenge obtaining compatibility with kinetic theory without the modification of the classical entropy current (or the kinetic theory).

**6.6. Other approaches.** There are many more methods of extending thermodynamics beyond local equilibrium. The test of these ideas usually starts with heat conduction. One may introduce the time derivatives of the state variables as additional state variables. However, time derivatives are frame dependent, they are many of them and it is not clear which one is to be used. Rigorous exploitation methods of the entropy principle do not help in this respect [85, 86]. Recently Serdyukov applied this idea for heat conduction, too [87, 88]. The ballistic-diffusive equation of Chen is a particular mixture of phenomenological and kinetic considerations [89, 90], the thermomass theory of Guo and Hou seemingly rediscovers the role of inertia in heat conduction [66].

An important class of heat conduction equations is nonlinear by construction [84, 66, 39, 67]. Possible experimental verifications require their reliable numerical solution. Analytical results are crucial in this respect [91, 92].

These approaches are partially consistent with either kinetic theory or continuum mechanics. They may use or avoid the usage of rigorous entropy principle exploitation. Some of them, like the delayed differential equations of dual phase lag theory [93] failed to fulfill important thermodynamic expectations [94, 95]. None of them were tested with experiments of ballistic-wave like-diffusive propagation.

## 7. DISCUSSION

In his mind provoking article Muschik writes that the reason of so many schools of thermodynamics is, that one may go beyond local equilibrium with different ways. The extension to nonlocal-nonequilibrium is not unique [96]. In this short survey we argued that the different extensions are not equivalent regarding their performance of modeling experimental results, neither regarding their theoretical consistency and scope.

The challenge of non-Fourier heat conduction is to develop a theory that is compatible both with kinetic theory and mechanical principles (including material frame indifference), passes the tests of existing experiments on parallel ballistic, wave like and diffusive propagation of heat and, therefore and most importantly, predictive in foretelling new observations and phenomena.

This pointed review is far from being complete and the author apologizes of not mentioning relevant works. This is partially because of his special point of view, beyond his limited knowledge.

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