Investigation and modelling of ultra-fast processes of energy transport : super-transport of energy

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NANYANG TECHNOLOGICAL UNIVERSITY

**INVESTIGATION AND MODELLING OF
ULTRA-FAST PROCESSES OF ENERGY
TRANSPORT: SUPER-TRANSPORT OF ENERGY**

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**SCHOOL OF MECHANICAL AND AEROSPACE
ENGINEERING**

2013

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BY

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ABSTRACT

This work is devoted to an investigation of ultra-fast processes of energy transport. There is a close link between ultra-fast and nano-scale processes. However, modern science lacks understanding of energy transport phenomena, which occurs at nano-space and time scales. In fact, a realistic model of such processes does not exist.

It was recently found that the phase-lagging models used to model ultra-fast processes lead to the existence of superfluid regime without low temperature condition. Still, the existing models are phenomenological and the kinetic theory cannot be used to describe these processes in any medium due to its limitations.

The research is focused on the existence of the fundamental time lag problem in the processes of heat transport. Quantum theory is a completely new approach in this area. It is used to solve the problem of the time lag existence and to estimate its value. It is proved in this thesis that the macroscopic equation for the second sound follows from the quantum equation for de Broglie's wave packet. It was shown that the result is consistent with the outcomes obtained previously from different theories. The investigation of the behavior of the apparent thermal diffusivity and viscosity was conducted as well as a study of the steady state heat transfer within a nano-scale spatial domain and apparent sources. The experiment was conducted to validate the resulting mathematical model, which appears to be in good agreement with the data obtained.

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Nomenclature

Latin Symbols

A	wave amplitude [m]
A^*	parameter
B	wave packet amplitude [m]
$B_{1,2}$	parameters
$B(t - t_0)$	instrument response function
C	arbitrary constant
C_p	specific heat [$\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$]
C_e	electron heat capacity [$\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$]
C_{ph}	phonon heat capacity [$\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$]
$C(r)$	arbitrary parameter function
$C_1(r)$	arbitrary parameter function
$C_2(r)$	arbitrary parameter function
$C(s)$	arbitrary parameter function
$C_1(s)$	arbitrary parameter function
$C_2(s)$	arbitrary parameter function
D	diffusion coefficient [$\text{m}^2 \cdot \text{s}^{-1}$]
D^*	apparent source parameter
$D(\varepsilon)$	density of states

E	energy [J]
F	laser irradiation fluence [$\text{J}\cdot\text{m}^{-2}$]
$G(T_e)$	electron-phonon coupling factor
$H(x)$	heat transfer coefficient [$\text{W}\cdot\text{K}^{-1}\cdot\text{m}^{-2}$]
$\bar{H}(\eta)$	heat transfer coefficient in the dimensionless form
H_{upper}	upper limit of the dimensionless heat transfer coefficient
I_{cl}	classical moment of inertia [$\text{kg}\cdot\text{m}^2$]
$I(t)$	light intensity of the laser pulse [$\text{W}\cdot\text{m}^{-2}$]
$I_0(z)$	modified Bessel function of the first kind
$I_1(z)$	modified Bessel function of the first kind of the first order
J	mass current [$\text{kg}\cdot\text{s}$]
$J(f)$	collision term
K_w	kernel $I_0(z)\exp(-z)$
$K(z)$	kernel $\frac{1}{\sqrt{\pi z}}$
$K_1(\eta)$	kernel $I_1\left(\frac{\eta}{2}\right)$
N_0	number of particles in a condensate
N	number of particles in a gas
$P(\eta, s)$	particular solution of the equation (6.2.12)
Pr	Prandtl number
$\tilde{Q}(\eta, s)$	Laplace transform of heat flux

R	radius [m]
R'	reflectivity
S	cross-section area [m ²]
$S(*,*)$	source term [K]
T	temperature [K]
$\tilde{T}$	Galilean transformed temperature [K]
T_0	initial temperature [K]
T_o	average temperature [K]
T_e	electron gas temperature [K]
T_p	phonon gas temperature [K]
T_λ	lambda-point [K]
U	potential energy [J]
U_e	excess velocity [m·s ⁻¹]
V	volume [m ³]
$V_{at}(r)$	true interatomic potential [m ² ·kg·s ⁻³ ·A ⁻¹]
$\boldsymbol{W}$	velocity gradient [s ⁻¹]
a	real variable
a_s	s-wave scattering length [m]
b	mean value of the normal distribution [s]
c	velocity [m·s ⁻¹]
d	thickness [m]

$\text{erf}(\xi)$	Gauss error function
f	distribution function
f_0	local equilibrium function
g	arbitrary function
h	Planck's constant [$\text{m}^2 \cdot \text{kg} \cdot \text{s}^{-1}$]
i	imaginary unit $\sqrt{-1}$
j	arbitrary function
k	wave number [m^{-1}]
$\mathbf{k}$	conductivity tensor [$\text{W} \cdot \text{K}^{-1} \cdot \text{m}^{-1}$]
$\bar{k}$	dimensionless thermal conductivity
k_B	Boltzmann constant [$\text{m}^2 \cdot \text{kg} \cdot \text{s}^{-2} \cdot \text{K}^{-1}$]
k_c	thermal conductivity [$\text{W} \cdot \text{K}^{-1} \cdot \text{m}^{-1}$]
k_{eff}	effective thermal conductivity [$\text{W} \cdot \text{K}^{-1} \cdot \text{m}^{-1}$]
k_e	electron thermal conductivity [$\text{W} \cdot \text{K}^{-1} \cdot \text{m}^{-1}$]
k_{ph}	phonon thermal conductivity [$\text{W} \cdot \text{K}^{-1} \cdot \text{m}^{-1}$]
k_w	relative wave factor [$\text{m}^{-1} \cdot \text{s}^{-1}$]
l	relative angular momentum [$\text{m}^2 \cdot \text{kg} \cdot \text{s}^{-2}$]
l_B	real constant
l_0	space quantum [m]
m	mass [kg]
m_r	reduced mass [kg]

p	pressure [Pa]
$p(x,t)$	specific solution [K]
$\boldsymbol{p}$	momentum, impulse [$\text{kg}\cdot\text{m}\cdot\text{s}^{-2}$]
q''	heat flux [$\text{W}\cdot\text{m}^{-2}$]
q''_{ph}	heat flux of the phonon gas [$\text{W}\cdot\text{m}^{-2}$]
q_v	volumetric source/sink term [$\text{K}\cdot\text{s}^{-1}$]
$\tilde{q}$	dimensionless volumetric heat source
$\boldsymbol{r}$	radius vector [m]
r_i	spatial coordinates
s	shear stress [Pa]
$s_{1,2}$	parameters
t	time [s]
t_p	full-width-half-maximum of the laser pulse [s]
u	medium velocity [$\text{m}\cdot\text{s}^{-1}$]
u_2	second sound velocity [$\text{m}\cdot\text{s}^{-1}$]
$\boldsymbol{v}$	velocity [$\text{m}\cdot\text{s}^{-1}$]
v_s	superfluid velocity [$\text{m}\cdot\text{s}^{-1}$]
w	arbitrary function
z	dimensionless variable
$\bar{z}$	dimensionless variable

Greek Symbols

Δ	energy of normal state of fluid [J]
$\Delta R'(t)$	differential reflection function
$\Delta R'_i$	amplitude of the component
$\theta(\eta, s)$	Laplace transform of temperature
α	hyperfine index; thermal diffusivity [$\text{m}^2 \cdot \text{s}^{-1}$]
α_{eff}	apparent thermal diffusivity [$\text{m}^2 \cdot \text{s}^{-1}$]
α_{ph}	thermal diffusivity of the phonon gas [$\text{m}^2 \cdot \text{s}^{-1}$]
$\check{\alpha}$	scalar value
$\bar{\alpha}$	dimensionless variable
β	the effective flux [$\text{kg} \cdot \text{s}^{-3}$]
$\check{\beta}$	scalar value
γ	real parameter
$\check{\gamma}$	scalar value
δ	characteristic length traveled by thermal waves [m]
δ^*	wave travelling distance [m]
$\delta(\xi)$	Dirac delta function
ε	kinetic energy [J]
ζ	real variable
η	dimensionless variable

θ	dimensionless temperature
θ'	real variable
$\theta_f(\xi)$	first integral in the expression (6.2.20)
$\theta_v(\xi)$	second integral in the expression (6.2.20)
ϑ	timelike variable
κ	radiation penetration depth [m]
λ	second viscosity [$\text{N}\cdot\text{s}\cdot\text{m}^{-2}$]
$\lambda(s)$	constant of integration
μ	dynamic viscosity [$\text{N}\cdot\text{s}\cdot\text{m}^{-2}$]
ν	frequency [s^{-1}];
ν_k	kinematic viscosity [$\text{N s}\cdot\text{m}^{-2}$]
ν_{eff}	effective viscosity [$\text{N}\cdot\text{s}\cdot\text{m}^{-2}$]
$\check{\nu}$	scalar value
ζ	real variable
ξ_b	real variable
ξ_p	real variable
π	constant
ρ	density [$\text{kg}\cdot\text{m}^{-3}$]
ρ_n	“normal” density [$\text{kg}\cdot\text{m}^{-3}$]
ρ_s	“superfluid” density [$\text{kg}\cdot\text{m}^{-3}$]
τ	time lag [s]

τ_{e-ph}	relaxation time corresponding to electron–phonon scattering [s]
τ_i	lifetime of the component [s]
τ_α	relaxation time due to thermal effects [s]
τ_ν	relaxation time due to viscous effects [s]
φ	energy flux [$\text{W} \cdot \text{m}^{-2}$]
ψ_0	condensate wave function
ψ	wave function
ω	frequency; angular velocity [s^{-1}]
ω_c	critical angular velocity [s^{-1}]

Special Symbols

$\hbar$	reduced Planck's constant [$\text{m}^2 \cdot \text{kg} \cdot \text{s}^{-1}$]
$\mathcal{U}$	Laplace transform of the excess velocity
$\otimes$	dyadic product
∇	nabla operator
∇^2	Laplace operator
$ \dots $	magnitude operator
Δx	uncertainty of the position [m]
Δp	uncertainty of the momentum [$\text{kg} \cdot \text{m} \cdot \text{s}^{-1}$]
$\langle , \rangle$	inner product

Abbreviations

BCS	Bardeen, Cooper and Schrieffer
BEC	Bose-Einstein condensation
FH	flying height (control)
FWHM	full-width-half-maximum
GMR	giant magnetoresistive head
GTP	Glan-Thompson Polarizer
HAMR	heat assisted magnetic recording
HDI	head-disk interface
IBM	international business machines
MQWs	multiple quantum wells
MRI	magnetic resonance imaging
OPO	Optical Parametric Oscillator
RTP	rapid thermal processing
SPL	single-phase-lagging
SQUIDs	superconducting quantum interface devices
STM	scanning tunneling microscope
TTM	two-temperature model
WLC	White Light Continuum

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Chapter 1. Introduction

It is highly important to investigate ultra-fast processes of energy transport as their significance cannot be overestimated. Modern technology moves towards even smaller scales and faster processes to improve functionality of devices we have right now. The key to breakthrough in this area is to understand and simulate the small-scale processes, such as nano-scale ones, to fabricate faster and smaller devices.

The investigation of nano-scale processes is at the forefront of modern science and technology. The main challenge and constraint is our lack of understanding of energy transport processes, which happen at nano-scale spacing and nano-scale time. In fact, a realistic model of such phenomena does not exist. Due to the lack of such a fundamental understanding, the existing models are phenomenological. Hence, new physical models are required to describe and successfully exploit properties of materials and processes at nano-scales.

For example, common electronics presently includes 10 nm components operating within nanosecond characteristic times. These kinds of devices are all around us: from home appliances to personal computers. Energy transfer is one of the most important aspects of electronic devices operation, which now takes place over nano-scale distances.

Why is macroscopic approach not applicable in this situation? The answer is simple. If we try to divide some substance into smaller and smaller pieces, then at some stage we would reach molecular and then atomic level, where the physical and chemical properties of the materials are completely different from those of the original piece of matter. Most biological processes, for example, occur at the molecular level. Many novel physical phenomena take place at the length scale of few nanometres. Carbon nanotubes with diameters of 0.4 nm and 50 nm have completely different properties.

In classical fluid mechanics and heat transfer, medium is treated as a continuum, assuming it can be divided infinitely without changing its nature. This model is not applicable in many current important situations and needs some alternative for a reliable description of different phenomena.

The appearance of quantum theory gives a lot of possibilities to develop instruments for exploring nano-scale phenomena. They are widely used in different fields of knowledge and industry. Some of these areas are introduced below [1].

1.1 Microelectronics and Information Technology

The progress in microelectronics is impossible without material sciences development, which are essential for crystal growing and thermal processing during semiconductor manufacturing. Rapid thermal processing (RTP) is necessary during annealing and oxidation to prevent ions from deep diffusion into the wafer. Thermal modeling of RTP

must consider the effects appearing due to conduction, convection and radiation. A better understanding of light scattering by anisotropic rough surfaces is also necessary as in some cases the wafer surface is rough and possesses anisotropic features.

The corresponding sizes of the components under investigation are about 20 nm or so. Optical energy is absorbed within milliseconds, so thermal diffusion simply does not have any time to occur to distribute heat uniformly across the wafer surface. To deal with these and many other problems, we must use molecular nanoelectronics together with quantum computing, which can brighten the electronics and computer future.

1.2 Lasers, Optoelectronics, and Nanophotonics

Lasers are widely used in modern technology. Furthermore, it is hard to imagine modern science without them. They are irreplaceable in metrology, microelectronics fabrication, manufacturing, medicine and communication.

The development of so-called quantum well lasers enables us to grow highly pure semiconductor thin films with atomic precision, due to their operation level of the discrete energy levels and enhanced density of states. They also have better performance with a smaller driving current. Multiple quantum wells (MQWs), also called superlattices, can be used to improve the performance too. Further improvement of laser efficiency has been made with the use of quantum wires and quantum dots.

Nanophotonics integrates in itself photonics with physics, chemistry, biology, materials science, manufacture and nanotechnology, which are far beyond macroscale level. Nanophotonics studies interactions between light and matter, investigates the properties of the nanostructures to utilise light energy, develops novel fabrications and sensing techniques. Some of the main goals of the research in this area are quantum electrodynamics, quantum dot and quantum wire lasers, etc., which all require a better understanding of the heat transfer processes on the micro and nanolevel.

1.3 Microfabrication and Nanofabrication

Microscopic devices are coming rapidly and strongly into our life. Even in 1983, Feynman [2] mentioned the use of the swimming machine (mobile microrobot) as a medical device: sort of electronic surgeon that you can swallow. Nowadays this is no longer a theory as microelectronics has tremendous success in commercial use, for example, micromachined accelerometers in the airbags. Microfabrication is used for medical and biological diagnostics, such as so-called lab-on-chip, with pump, valve and analysis sections on the 10 to 100 μm scale. In aerospace engineering, it is used for building micro-air vehicles or microflyers that could be used under extreme conditions. Microchannels and microscale heat pipes are developed for cooling electronic devices.

One of the successful technologies that operate in the regime of quantum mechanical domain is the giant magnetoresistive (GMR) head and hard drive. This technology was

first introduced by IBM in 1996, only ten years after the original research [3, 4]. GMR materials have been extensively used in computer hard drive and read/write head.

In all these devices, quantum behaviour of the medium becomes so important that the effects of the quantum theory cannot be neglected. Hence, quantum mechanics is inevitable for understanding the behaviour of the matter at these scales.

1.4 Probing and Manipulation of Small Structures

One of the most peculiar quantum mechanical phenomena is tunnelling by elementary particles, which refers to their wavelike behaviour. It stands for a potential barrier that normally will confine the particles to either side of the barrier. This can be represented by a mountain, that is so high and wide, that separates people on one side from those on the other. When the barrier thickness is small, the effect of quantum tunnelling can occur and particles are transmitted through the barrier, like a person who would appear on the other side of the mountain suddenly.

This effect is used in a device, called Josephson junction, which is used in superconducting quantum interface devices (SQUIDs), for measuring extremely small magnetic fields. SQUIDs are also used in magnetic resonance imaging (MRI) for medical diagnostics. Scanning tunnelling microscope (STM) is also based on this phenomenon (electron tunnelling through vacuum). This device has enabled the detection and manipulation of surface phenomena at the atomic level and, thus, has enhanced the

applications of nanoscale science and technology. Other types of STMs are developed today, which help in such areas of research as friction measurements, nanoscale indentation, dip-pen nanolithography, etc.

1.5 Energy Conversion Devices

Nanostructures may have unique thermal properties which can be used for heat removal and thermal management applications. For example, nanotubes can be used for cooling of computational devices. Nanofluids also can be used for these purposes, which are of extreme importance now. Nanofluids are liquids with suspensions of nanostructured solid materials, such as nanoparticles, nanofibres and nanotubes. They provide enhanced thermal conductivity and increased thermal flux, but the mechanisms of these properties are still under investigation [5-8].

Nanostructures are used to manufacture surfaces with specified absorption, reflection and emission characteristics. Moreover, at the nanoscale, the radiative energy transfer can be greatly enhanced due to tunnelling and enhanced density of states. Nanostructures can also help increase the energy conversion efficiency and reduce the cost of solar cells. For example, Nanosolar achieved more than 16% efficiency of solar energy conversion with their nano foil cells [9].

Nanomaterials are being developed for several key issues related to hydrogen technologies, such as hydrogen storage using nanoporous materials, effective hydrogen

generation by harvesting solar energy with inexpensive photovoltaic materials, and fuel cells based on nanostructure catalysts.

1.6 Biomolecule Imaging and Molecular Electronics

Optical microscopy is of great importance in medical diagnoses because it allows us to see bacteria and blood cells. Optical wavelengths are more desired for these purposes, than X-rays, because they are less invasive and more convenient. However, the resolution of the traditional microscope does not allow the observation of small objects due to the diffraction limit. Many different technologies, like near-field scanning optical microscopes are used for these purposes nowadays, which all require nanoscale fabrication.

Molecular electronics is considered a promising perspective of modern technology. A three dimensional assembly with short interconnect distances would greatly increase the information storage density and transfer speed with reduced power consumption and amount of heat dissipated. Because of the small dimensions, quantum mechanics should govern the electrical and mechanical behaviours as a branch of Physics which describes phenomena happening at very small scales (action is on the order of the Planck constant). Molecular electronics is in its infancy now, but it is expected to make a revolution in the electronics industry in this century [10, 11].

All these issues are not possible without theoretical investigations, as engineers create technological advances using theoretical findings, and one is impossible without the other. In this project, we focus on the theoretical aspects of heat transfer to deal with the inaccuracies of previous theories, which are of great importance in modern science and its applications.

In our case, classical assumptions made for energy transport at macroscales, such as Fourier law, are no longer applicable as the continuum hypothesis fails at these scales and physical laws governing these phenomena take different mathematical forms.

Although the classical Fourier law leads to the paradox of instantaneous energy propagation and infinite speed of propagation, it works well at the macro-level, but it no longer can be used to model ultra-fast processes: other constitutive equations are to be employed. To deal with this paradox, the phase-lagging models have been proposed [12, 13]. The analysis of the solutions to the phase-lagging energy equations suggests that when the characteristic time of the process is much smaller than the lag time, the wave mode of transport becomes the main mechanism of energy transfer [14].

In ultra-short heat transfer processes, the wave transport is manifested by an apparent increase (three-four orders of magnitude) of the thermal diffusivity of the transporting medium [15]. In viscous fluid flow, in addition, an apparent decrease of viscosity occurs, so that the liquid behaves as a superfluid even at ambient temperature and pressure [16].

It has been demonstrated that the lag time is inversely proportional to the square root of temperature [15]. Thus the life span of the super-transport phenomena should significantly increase at lower temperatures, which is consistent with the observation that certain fluids behaved as superfluids at low temperatures for long periods of time.

From the point of view of the quantum physics, the waves transporting energy can be viewed as narrow wave packets, whose wave functions are able to describe the superfluid state of the liquid. This may provide an alternative explanation of the phenomenon of superfluidity, i.e. the Bose-Einstein condensation may not be needed for superfluidity to occur [17].

Theoretical predictions show that the life span of this phenomenon is extremely short: tens or hundreds of pico-seconds, which then appears to be an ultra-fast process. If harnessed, however, the phenomenon of high-temperature superfluidity may become an excellent – and, perhaps, the only tool – for providing efficient (as the liquid behaves as inviscid) cooling of micro-electronic and nano-devices as well as quantum computers (because of an apparent increase - three-four orders of magnitude - of the thermal conductivity of the coolant).

Based on the recent theoretical developments and computer simulations [18, 19], it seems feasible to design a prototype system: a nano-channel or a maze of such channels, or a porous medium composed of nano-sized pores, in which fluid flow would be generated by high-frequency pulses (10-100 GHz), so that the superfluid flow regime could be

maintained for a long span of time. An additional enhancement can be achieved by choosing coolants with a larger value of the relaxation (lag) time. Some change of physical conditions (i.e. pressure change) may also lead to the increase of the lag-time.

In addition, the phenomenon of high-temperature superfluidity can be used for enabling super-transport of energy, that is, transporting huge energy packets within ultra-short spans of time. To achieve this, it is necessary to demonstrate that the amount of energy necessary to maintain the superfluid state is much less than the transported amount. This requires additional investigations.

The above project is a major step towards developing a cooling system for micro-electronic and nano-devices, as well as quantum computers, which has no equivalent in the world. The resulting system will be patented with its following commercialisation.

1.7 Objectives and Scope of the Present Study

Some theoretical issues of the model are still to be understood and investigated before one can proceed with practical implementations.

The resulting model may be applied, in general, to all circumstances, where heat transfer occurs at nano-scales or during ultra-fast processes. The goal of this study is to achieve understanding of processes at nanoscale level. The objectives of this thesis are:

- To acquire a fundamental understanding of the physical processes at nano-scales and ultra-fast transport processes;
- To develop a realistic model of energy transport processes at nano-scale, ultra-fast transport processes and tools to simulate such processes;

Improvement of cooling of electronic devices, enhancement of efficiency of solar cells cannot be established before developing a theoretical background of the underlying phenomena (for possible practical implementations of this study see pages 1-10 of the thesis). Investigation of these issues is the scope of this project, namely:

- It is conjectured that phenomenological phase-lagging formulation of ultra-fast transport processes can be rigorously derived from the quantum theory, as existing models are proved to be inconsistent in general case due to paradox of instantaneous propagation. This derivation is the first task of this project.
- The phase-lagging transport equations appear to have an apparent source/sink term in them, the physical meaning of which is still not clear. Another task of the thesis is to determine its significance and investigate the properties of these apparent source/sink terms.

- The lag time is a function of thermodynamic variables (e.g., temperature, pressure) as well as the values of the physical properties of the transporting medium. The task is to rigorously obtain this function from the principles of statistical physics and quantum theory.
- Investigate and model any other effects related to the ultra-fast energy transport (e.g., quantum effects, relativistic effects - if any, - etc.) is the final task of the present study.

These tasks result in the following deliverables:

- Fundamental understanding of the energy transport processes at nano-scales.
- A novel (realistic) mathematical model of the energy transport processes (nano-scale & ultra-fast) derived from the quantum theory standpoint.
- Tools to simulate such processes numerically.
- Proposals of harnessing energy super-transport states.

Possible applications of this study may result in:

- the development of thermal sensing mechanism and technology for contact detection;
- thermal analysis and management in heat transfer across head-disk interface (HDI) of heat assisted magnetic recording (HAMR);
- thermal actuation analysis for flying height (FH) control;
- other energy super-transport phenomena in nano-scale channels.

The phenomenon of high-temperature superfluidity, which is one of the main focuses of this project, if harnessed, can be used for enabling super-transport of energy, that is, transporting huge energy packets within ultra-short time spans.

1.8 Thesis Overview

This study focuses on the investigation and modelling of ultra-fast processes of energy transport. In this thesis, the introduction (Chapter 1) discusses the present-day state of science and technology and motivation of this study, followed by the objectives and scope. Chapter 2 provides a detailed discussion on the fundamental time lag existence in heat transfer processes, history of the lagging heat transfer investigations, attempts to solve the problem and present condition of the study. It is followed by a brief history of quantum theory and its connections to the macroscopic problems. Furthermore, Chapter 2

describes the superfluidity phenomenon and its up-to-date state as well as several still unsolved problems. Chapter 3 illustrates the author's solution of the time lag existence problem and its estimation with the quantum theory approach. It is proven that this solution is consistent with the results obtained previously from different theories. In Chapter 4, a phenomenon of apparent thermal diffusivity and viscosity is discussed as well as a steady state heat transfer problem. Chapter 5 is dedicated to the investigation of the apparent source/sink term and its role on the flow affected by laser pulse (Gaussian distribution). Chapter 6 presents experimental results and model validation. And finally, Chapter 7 provides overall results of the study as well as a summary of findings, discussion on their physical significance and ways of their experimental and commercial applications.

Chapter 2. Literature Review

2.1 Problem Hidden Behind the Fourier Law

2.1.1 Introduction

The classical linear heat equation is derived from the energy conservation equation [20]:

$$\rho c \frac{\partial T}{\partial t}(x, t) + \nabla \mathbf{q} \cdot (x, t) = S(x, t, T), x > 0, t > 0 \quad (2.1.1)$$

where $S(x, t, T)$ is the volumetric source/sink term, T denotes temperature and $\mathbf{q}$ represents the local heat flux. This relation contains two unknowns, that is why the constitutive relation is needed. If the Fourier law $\mathbf{q} = -k_c \nabla T$ is used, then the heat equation follows:

$$\frac{\partial T}{\partial t} = \alpha \nabla^2 T(x, t) + \frac{1}{\rho c} S(x, t, T), x > 0, t > 0 \quad (2.1.2)$$

where k_c is the thermal conductivity, $\alpha = \frac{k_c}{\rho C_p}$ represents the thermal diffusivity, ρ denotes density and C_p stands for specific heat at a constant pressure.

The global solution for the equation (2.1.2) is [20]

$$T(x, t) = \frac{1}{\sqrt{4\pi\alpha t}} \int_0^\infty T(x, 0) e^{-\frac{x^2}{4\alpha t}} dx, \quad (2.1.3)$$

$$\forall t, x \quad T(x, 0) \neq 0 \text{ since } e^{-\frac{x^2}{4\alpha t}} \neq 0 \quad \forall t, x$$

Hence, from equation (2.1.3), the paradox of instantaneous propagation of energy follows. However, in classical problems this paradox is not manifested, but it becomes inevitable as $x \sim \sqrt{\alpha t}$. It means that ultra-fast and nano-scale processes cannot be modelled by the classical heat equation.

In order to deal with this paradox, it is assumed that the flux and the gradient occur at different moments of time [20]: the flux then starts at a later time, then:

$$\mathbf{q}(x, t + \tau) = -k_c \nabla T \quad (2.1.4)$$

Expanding this relation into Taylor series and neglecting the terms of expansion with orders larger than one upon the condition that τ is small (which can be assumed from empirical experience), we obtain the following expression:

$$\tau \frac{\partial \mathbf{q}}{\partial t} + \mathbf{q} = -k_c \nabla T \quad (2.1.5)$$

This equation was first introduced by Maxwell [21], who cast out the time derivative with the remark that it “... may be neglected, as the rate of conduction $\mathbf{q}$ will rapidly establish itself.” So, Maxwell has never performed the analysis of short-term effects and did not note that diffusion is associated with an infinite speed of propagation.

50 years later, in 1917, Nernst [22] proposed that in good thermal conductors at low temperatures heat could have sufficient “inertia” to give rise to oscillatory discharge. Later, in 1935 the paradox was first addressed in heat transfer by Davydov [23].

In 1938, Tisza discovered superfluid helium II [24] and derived a wave equation for heat and a formula that predicts extremely small wave speeds for the propagation of heat in liquid helium II. Landau later called this heat wave a “second sound” and developed the two-fluid theory for liquid helium II [25]. From the two speeds he found, one stands for the ordinary sound and the other – for a second sound, which describes the propagation of the temperature waves. But propagation of the Landau waves in liquid helium II is of quantum nature and does not mean that in ordinary materials the heat transport should

occur by wave propagation. The liquid helium in Landau's work was represented by a phonon gas of elementary excitations near absolute zero. As the compressional sound wave propagates through the phonon gas, the periodic variations of phonons density occur due to temperature variations in liquid helium, that is, second sound. It seems that Landau considered that heat propagation was a special phenomenon related to phonon excitation; and, there is no mention of the paradox of heat diffusion in Landau's works.

2.1.2 Cattaneo's Model

The first to propose an explicit mathematical theory to correct unacceptable properties of the Fourier theory of heat diffusion was Cattaneo [26]. He used the kinetic theory and a second-order correction of this to derive the expression for the flow $\mathbf{q}$ in one dimensional space:

$$\tau \frac{\partial \mathbf{q}}{\partial t} = -\mathbf{q} - k_c \frac{\partial T}{\partial x} \quad (2.1.6)$$

where T is the temperature, k_c is the thermal conductivity and τ represents the relaxation time.

With the one-dimensional energy equation

$$\rho C_p \frac{\partial T}{\partial t} = \frac{\partial q}{\partial x} \quad (2.1.7)$$

where C_p is the specific heat of gas at a constant pressure, it follows that the temperature T and the heat flux satisfy the same telegraph equation:

$$\tau \frac{\partial^2 T}{\partial t^2} + \frac{k_c}{\rho C_p} \frac{\partial^2 T}{\partial x^2} + \frac{\partial T}{\partial t} = 0 \quad (2.1.8)$$

The propagation speed due to this equation is $c = \sqrt{\frac{k_c}{\rho C_p \tau}}$.

Cattaneo refers neither to the work of Maxwell nor to other works, such as the one by Landau, in which second sound is identified as the wave propagation of heat. Cattaneo narrowed his research to gases and did not consider heat propagation in liquids or solids.

2.1.3 Further Studies

In 1951, Ward and Wilks again derived the Landau's expression for the second sound [27] directly from a phonon gas model without referring to the two-fluid theory. It is of particular significance because, as phonon gas excitations exist in solids and some liquids, the second sound then should be detectable in solids and liquids like in helium II.

A year later, they derived a wave equation for the second sound [28]:

$$\frac{\partial^2 E}{\partial t^2} = \frac{c_1^2}{3} \frac{\partial^2 E}{\partial x_i \partial x_i} \quad (2.1.9)$$

where E is the momentum of the localized phonons, and c_1 is the speed of sound. It is assumed that all phonons travel at a constant speed c_1 . Starting with the Boltzmann equation, the authors make an assumption that there are many phonon collisions in a distance equal to the wavelength of any disturbance that may be propagated.

In 1953, Morse and Feshbach proposed a hyperbolic equation of heat conduction. However, they did not give any rigorous proof for it [29].

2.1.4 Cattaneo-Vernotte Model and Tavernier's Derivation

In 1958, Vernotte postulated a rate equation for the heat flux and derived a telegraph equation for the temperature [30] and in 1961 found an inconsistency in Cattaneo's derivation of the heat law because the second-order correction of the Maxwell's distribution was ignored [31].

Using the Boltzmann's equation

$$\frac{\partial f}{\partial t} + \mathbf{v} \text{grad} f = \left. \frac{\partial f}{\partial t} \right|_{\text{coll}} \quad (2.1.10)$$

Tavernier [32] obtained the rate equation

$$\mathbf{q} = -\mathbf{k} \text{grad} T - \tau \frac{\partial \mathbf{q}}{\partial t} \quad (2.1.11)$$

where $\mathbf{k}$ is a conductivity tensor, τ is the mean relaxation time, and f denotes the partition function in the absence of the external forces. The collision term $\left. \frac{\partial f}{\partial t} \right|_{\text{coll}}$ is approximated by

$$-\frac{f - f_0}{\tau[\mathbf{v}]} \quad (2.1.12)$$

where f_0 is the equilibrium distribution which corresponds to the temperature at the point and τ is the relaxation term for the particles with velocity $\mathbf{v}$. He also showed the derivation of the rate equation for solids where the energy transport can be carried out by two types of particles: phonons and electrons. Finally, the rate equation leads to the telegraph equation and a single speed for heat waves.

The first paper to review works on the hyperbolic heat equation and second sound was written by Chester [33]. There he postulated the Cattaneo equation and said that the second sound can appear in any material which can be modelled as a phonon gas. He stated that wave propagation is important when

$$\left| \frac{\partial T}{\partial t} \right| \gg \left| \frac{T}{\tau} \right| \quad (2.1.13)$$

and, in case of a reversed inequality, the diffusion dominates. This criterion can be expressed through a critical frequency:

$$\omega_c = \frac{1}{2\pi\tau} \quad (2.1.14)$$

which stands for the diffusion for the lower frequencies and waves for higher ones. An expression of τ in terms of the sound speed was also obtained.

2.1.5 First Experimental Measurements and Further Investigations

First successful measurement of second sound was done with a pulse technique in solid helium [34]. For heat pulse propagation in Al_2O_3 crystals, the telegraph equation may also be applied under certain assumptions [35].

At the same time, Guyer and Krumhansl [36] solved a linearized Boltzmann equation for a pure phonon field in terms of the eigenvectors of the normal-process collision operator. Steady one-dimensional solutions are derived for the cylinder of radius $r=R$ with $q(R)=0$. When the relaxation time for momentum-nonconverting processes is negligible, the umklapp processes (anharmonic phonon-phonon, or electron-phonon, scattering processes, creating a phonon with a momentum $\mathbf{k}$ -vector outside the first Brillouin zone - a uniquely defined primitive cell in reciprocal space) are negligible to normal ones, the solutions are reduced to the Poiseuille flow of the phonon gas. The formula for the thermal conductivity is derived to classify different regions of heat transport:

- ballistic – the region where the principal mechanism for momentum loss by the phonon gas is the direct flight of the phonons to the boundaries of the sample;
- Poiseuille flow – the region of the momentum loss by diffusion to the boundaries of the sample;

- Ziman – in this region the Poiseuille flow condition is no longer valid because of the increased scattering processes;
- kinetic – in this region the principal mechanism of the momentum loss continues to be the scattering distributed through the bulk sample [36, 37].

A relativistic generalisation of Cattaneo’s law was first proposed by Kranys [38], because signals should not propagate faster than the speed of light. As it was written, this requirement is not satisfied by the Fourier law. The Kranys system removes the paradox of an infinite velocity value, but his heat-flow equations may be not properly invariant (for observers in different points that move as rigid bodies) [39].

A theory of the propagation of macroscopic thermal quantities (local temperature, heat, phonon drift, heat capacity) in terms of the microscopic quantities of lattice dynamics was first proposed in the paper by Enz [40]. Two different temperature waves with two different speeds were identified. There are two second sounds, which are manifested under different conditions, defined by frequency and temperature. A drifting second sound seems to be observed in the most of the experiments, as the “driftless second sound” happens to occur only at high frequencies and the situation, where it can be observed, is associated with the heat conduction domination.

The theory of heat conduction by Gurtin and Pipkin [41] uses constitutive wave assumptions that lead to finite wave speeds and do not allow for an effective thermal

conductivity. The heat flux is said to be determined by the history of the temperature gradient, like in the models of viscoelastic materials with instantaneous elasticity. This instantaneous elastic part means that the heat flux is always finite, even at the initial moment where it produces a finite wave speed. The assumptions of the theory lead to the conclusion that the waves travelling in the direction of the heat flux vector propagate faster than the waves that travel in the opposite direction. The linearised equations for the heat flux and the internal energy have the form

$$\begin{aligned} \mathbf{q}(\mathbf{x}, t) &= - \int_{-\infty}^t Q(s) \nabla T(\mathbf{x}, t - s) ds, \\ \mathbf{e}(\mathbf{x}, t) &= b + C_p T(\mathbf{x}, t) + \int_0^\infty F(s) T(\mathbf{x}, t - s) ds \end{aligned} \tag{2.1.15}$$

where the kernels Q and F have a finite instantaneous value and b is constant. The internal energy functional $\mathbf{e}(\mathbf{x}, t)$ depending on the history of the temperature and the heat flux in a differentiable way is also introduced.

In 1969, Fox [42] developed a nonlinear constitutive theory of thermoelasticity. The material time derivative $d\mathbf{q}/dt$ is assumed to be dependent on the deformation gradient, $\mathbf{q}$, the temperature, T , and the temperature gradient, but Fox did not solve the invariance problem completely, and the problem of the invariant convective derivatives has many different solutions.

Maurer indicated [43] that Tavernier [32] used a linearized form of the Boltzmann equation neglecting quantum-mechanical effects and presented a time-dependent relaxation model for the heat flux in metals. The model is derived from the quantum-mechanical form of the Boltzmann transport equation under the assumption that phonons are in thermal equilibrium and, for treating electron-phonon interactions, the Lorenz approximation is used. The estimated relaxation time of 10^{-14} s in monovalent metals was obtained.

The Cattaneo equation was derived from the formulation of the thermodynamics of irreversible processes in the relativistic case by Müller [44].

The conditions for the existence of two types of second sound, “drifting” and “driftless”, were obtained from the exact solution of the complete linearized Boltzmann equation [45]. This solution is given in terms of eigenvalues and eigenvectors of the collision matrix including the effects produced by umklapp processes, normal processes and imperfections. The relaxation time for both processes appears to be the same and equal to the reciprocal of the smallest non-zero eigenvalue term of the collision matrix. Moreover, the analysis indicates three propagation speeds, instead of the two mentioned before, and the possibility of even other types of second sound is conjectured.

2.1.6 Time Lag for Solids

Later, Müller presented a thermodynamic theory for thermoelastic materials [46]. It shows finite speeds of propagation of temperature and a symmetric conductivity tensor.

The telegraph equation is derived for the heat conducted in a body of uniform density at rest.

However, Gurtin and Pipkin's equations (2.1.15) [41] were generalised by Nunziato [47] but, unfortunately, with no underlying physical reason. The generalisation was done by permitting the heat flux to depend on the current temperature gradient and on its history.

Nunziato derived the formulas describing the speed and attenuation of jumps in $\frac{\partial T}{\partial t}$.

Another generalisation [48] was obtained in thermoelasticity leading to a symmetric heat conduction tensor and Cattaneo's law by generalisation of entropy production inequality described by Müller [46].

Using the Cattaneo equation, the thermal stress in solids was calculated due to the sudden change in the heat flux [49]. It was showed that the correct model leads to high momentary temperature because of a step jump in the heat flux.

The three-dimensional case was studied by Tsai and MacDonald [50] and the methods of solution were almost the same as in the two-dimensional case. The lattice was formed by individual atoms and a set of equations of motion, obtained from an interatomic potential, was solved numerically.

In the study of the Boltzmann equation for a phonon gas, which models dielectric crystals with negligible electronic contribution, the constant thermal phonon relaxation time was

not assumed [51]. It was showed that in most cases of phonon interactions, $\frac{\tau}{k^n} = \text{const}$, where k is the wave number and $n \geq 1$. It was showed that the Cattaneo equation cannot yield a dispersion relation for harmonic waves that holds over a wide range of frequencies, because of the dependence of relaxation times on the phonon wave number. With the choice of the suitable memory kernel, the dispersion relations probably may hold over a wide range of frequencies. The idea of this work was not followed by anyone afterwards.

2.1.7 Back to the Cattaneo Equation

In 1976, the Cattaneo equation was obtained with the assumption that entropy depends on the temperature [52].

However, it was shown that the Cattaneo equation may be incorrect in the case when the heat carrier's time depends on the wave number [53]. It was proved by obtaining the solution for a plane wave as a Fourier transform and finding the solution for the heat flux from the energy balance. The dispersion relations [51] are used to invert the transform and to obtain the laws for heat flow for long waves but all this procedure never leads to the Cattaneo law or the telegraph equation, though interesting partial differential equations were obtained subject to the assumptions made about the wave number dependence on the phonon relaxation times.

Allowing the matrices of the relaxation times to depend on temperature and using some approximations, the nonlinear generalisation of the Cattaneo law has been obtained [54]:

$$\mathbf{T}(T) \frac{D\mathbf{q}}{Dt} + \mathbf{q} = -\mathbf{k}(T) \nabla T \quad (2.1.16)$$

where heat flux is $\mathbf{q} = \frac{c_p T_0}{3} \mathbf{v}$, $\mathbf{v}$ is the fluid velocity, and T_0 is an average temperature in the range of temperatures in which the second sound would propagate. Hence, we obtain:

$$\frac{D\mathbf{q}}{Dt} = \frac{\partial \mathbf{q}}{\partial t} + (\mathbf{v} \cdot \nabla) \mathbf{q} + \mathbf{W}[\mathbf{v}] \mathbf{q} \quad (2.1.17)$$

where $\mathbf{W}[\mathbf{v}]$ is the velocity gradient or the negative transpose of this gradient, or some linear combination of these [42]. Each invariant derivative leads to different constitutive laws, and it is impossible to know which one to choose.

In order to replace Cattaneo's theory, a new general nonlinear model for heat conduction in solids was proposed [55]:

$$(\check{\alpha}'_0 \mathbf{q}) + \nabla \cdot (\check{\gamma} \mathbf{1} + \check{\beta} \mathbf{q} \otimes \mathbf{q}) = -\check{\nu} \mathbf{q} \quad (2.1.18)$$

where $\otimes$ is a dyadic product and $\check{\alpha}, \check{\beta}, \check{\gamma}$ and $\check{\nu}$ are scalar values. This equation leads to the Cattaneo law when $\check{\beta} = 0$, $\check{\alpha} = \text{const}$ and $\check{\gamma}$ and $\check{\nu}$ are functions of T . It is assumed that the internal energy E and entropy depend on T and $\mathbf{q}$. The theory is consistent with thermodynamics as it uses an entropy inequality and the governing equations are:

$$E_\theta \dot{\theta} + E_w \mathbf{q} \cdot \dot{\mathbf{q}} + \nabla \cdot \mathbf{q} = 0 \quad (2.1.19)$$

$$\check{\alpha}'_0 \dot{T} \mathbf{q} + \check{\alpha}_0 \dot{\mathbf{q}} + \Phi'_0 \nabla \theta + \check{\nu} \mathbf{q} = 0 \quad (2.1.20)$$

This is a hyperbolic system and the equation (2.1.19) describes the energy balance. Authors believe that this model fits the experimental data pretty well. The difference between these equations and the Cattaneo equation is in the nonlinear term $\check{\alpha}'_0 \mathbf{q} \dot{T}$.

2.1.8 Galilean Principle of Relativity

Despite all these efforts, one of the best models today is the Cattaneo-Vernotte model. While it still describes the reality quite well, it suffers from one significant imperfection:

it violates the Galilean principle of relativity [56]. This is why it is necessary to find another model which satisfies relativity principle.

Let us consider the following Galilean transformation:

$$\begin{cases} \mathbf{r} = \mathbf{r}' - \mathbf{U}t \\ t = t \\ T(\mathbf{r}, t) = \tilde{T}(\mathbf{r}', t) \\ \mathbf{q}(\mathbf{r}, t) = \mathbf{q}'(\mathbf{r}', t) \end{cases} \quad (2.1.21)$$

where $\mathbf{U}$ is the constant velocity between two inertial reference frames, $\mathbf{r}$ is the radius vector in new coordinate system, $\mathbf{q}'$ and $\tilde{T}$ are $\mathbf{q}$ and T after Galilean transformation, respectively. There is no time shift in this transformation; hence, t remains the same. From these relations, it is clear that $\nabla_{\mathbf{r}} = \nabla_{\mathbf{r}'}$. Therefore, we have

$$\nabla_{\mathbf{r}'} T(\mathbf{r}', t) = \nabla_{\mathbf{r}} \tilde{T}(\mathbf{r}, t) \quad (2.1.22)$$

Hence, the Cattaneo's equation of a single-phase-lagging (SPL) heat conduction model [56] takes the following form:

$$\mathbf{q}'(\mathbf{r}', t + \tau) = -k_c \nabla_{\mathbf{r}'} T(\mathbf{r}', t) \quad (2.1.23)$$

It is clear that there is no $\mathbf{U}$ in this expression. Hence, the SPL model can be used in the moving media as it does not violate the Galilean relativity principle.

If we expand the left hand side into the first order Taylor series, it will lead to the Cattaneo-Vernotte's model which violates the above-named principle. The question is whether the Galilean principle will still be violated if we expand it into a higher order series. The second order approximation yields:

$$\mathbf{q}(\mathbf{r}, t) + \tau \frac{\partial \mathbf{q}(\mathbf{r}, t)}{\partial t} + \frac{\tau^2}{2} \frac{\partial^2 \mathbf{q}(\mathbf{r}, t)}{\partial t^2} = -k_c \nabla \tilde{T} \quad (2.1.24)$$

With the Galilean transformation, written before in equation (2.1.21), we obtain:

$$\frac{\partial \mathbf{q}(\mathbf{r}, t)}{\partial t} = \frac{\partial \mathbf{q}(\mathbf{r}', t)}{\partial t} - \frac{\partial \mathbf{q}(\mathbf{r}', t)}{\partial \mathbf{r}'} \cdot \mathbf{U} \quad (2.1.25)$$

$$\frac{\partial^2 \mathbf{q}(\mathbf{r}, t)}{\partial t^2} = \frac{\partial^2 \mathbf{q}(\mathbf{r}', t)}{\partial t^2} - 2 \frac{\partial^2 \mathbf{q}(\mathbf{r}', t)}{\partial t \partial \mathbf{r}'} \cdot \mathbf{U} + \mathbf{U} \cdot \frac{\partial^2 \mathbf{q}(\mathbf{r}', t)}{\partial \mathbf{r}'^2} \cdot \mathbf{U} \quad (2.1.26)$$

After substitution of these into the equation (2.1.24), we obtain:

$$\begin{aligned} \mathbf{q}'(\mathbf{r}', t) + \tau \left[\frac{\partial \mathbf{q}(\mathbf{r}', t)}{\partial t} - \frac{\partial \mathbf{q}(\mathbf{r}', t)}{\partial \mathbf{r}'} \cdot \mathbf{U} \right] \\ + \frac{\tau^2}{2} \left[\frac{\partial^2 \mathbf{q}(\mathbf{r}', t)}{\partial t^2} - 2 \frac{\partial^2 \mathbf{q}(\mathbf{r}', t)}{\partial t \partial \mathbf{r}'} \cdot \mathbf{U} + \mathbf{U} \cdot \frac{\partial^2 \mathbf{q}(\mathbf{r}', t)}{\partial \mathbf{r}'^2} \cdot \mathbf{U} \right] \\ = -k_c \nabla_{\mathbf{r}'} \tilde{T}(\mathbf{r}', t) \end{aligned} \quad (2.1.27)$$

It can be easily seen that this equation violates the Galilean principle of relativity. Similar deduction leads to the conclusion that the higher order approximations will have the same problem. However, a good model must be also valid in a moving medium, that is why it is necessary to find a model that satisfies Galilean principle of relativity.

2.1.9 Modern Single-Phase-Lag Model

An elegant derivation of the single-phase-lag [SPL] model from the Boltzmann equation was first performed by Tavernier [32] and repeated by Lin et al. [56]. In the absence of external forces, the Boltzmann equation takes the form:

$$\frac{\partial f(\mathbf{r}, \mathbf{v}, t)}{\partial t} + \mathbf{v} \cdot \nabla f(\mathbf{r}, \mathbf{v}, t) = \left(\frac{\delta f}{\delta t} \right)_{coll} \quad (2.1.28)$$

here $f(\mathbf{r}, \mathbf{v}, t)$ is the probability density of finding a classical particle at position $\mathbf{r}$, at a time t with the speed $\mathbf{v}$, $\left(\frac{\delta f}{\delta t} \right)_{coll}$ is the collision term which represents the rate of change of $f(\mathbf{r}, \mathbf{v}, t)$ due to the particle collisions. The particles here refer to the heat carriers such as electrons and phonons. Here it is assumed that initially the system is not at equilibrium described by a distribution function $f(\mathbf{r}, \mathbf{v}, t)$, which reaches a local equilibrium condition exponentially, as a result of collisions, with a relaxation time τ , described by a function $f_0(\mathbf{r}, \mathbf{v})$.

The collision term is approximated as follows:

$$\left(\frac{\delta f}{\delta t} \right)_{coll} = -\frac{(f - f_0)}{\tau} \quad (2.1.29)$$

The Boltzmann's equation then becomes:

$$\frac{\partial f}{\partial t} + \mathbf{v} \cdot \nabla f = -\frac{(f - f_0)}{\tau} \quad (2.1.30)$$

Using the definition of $f(\mathbf{r}, \mathbf{v}, t)$, the heat flux vector $\mathbf{q}$ can be expressed as [51]

$$\mathbf{q}(\mathbf{r}, t) = \int_{\varepsilon} \mathbf{v}(\mathbf{r}, t) f(\mathbf{r}, \varepsilon, t) \varepsilon D(\varepsilon) d\varepsilon \quad (2.1.31)$$

where ε is the kinetic energy of a particle, and $D(\varepsilon)$ is the density of states.

Using a first order difference scheme, we approximate the time derivative in the Boltzmann equation:

$$\begin{aligned} \frac{f(\mathbf{r}, \varepsilon(\mathbf{v}), t + \Delta t) - f(\mathbf{r}, \varepsilon(\mathbf{v}), t)}{\Delta t} + \mathbf{v} \nabla f(\mathbf{r}, \varepsilon(\mathbf{v}), t) \\ \cong -\frac{f(\mathbf{r}, \varepsilon(\mathbf{v}), t) - f_0(\mathbf{r}, \mathbf{v})}{\tau} \end{aligned} \quad (2.1.32)$$

Setting $\Delta t = \tau$, we obtain

$$\begin{aligned}
& \frac{f(\mathbf{r}, \varepsilon(\mathbf{v}), t + \tau) - f(\mathbf{r}, \varepsilon(\mathbf{v}), t)}{\tau} + \mathbf{v} \nabla f(\mathbf{r}, \varepsilon(\mathbf{v}), t) \\
& \cong - \frac{f(\mathbf{r}, \varepsilon(\mathbf{v}), t) - f_0(\mathbf{r}, \mathbf{v})}{\tau}
\end{aligned} \tag{2.1.33}$$

Upon rearranging the terms in the equation (2.1.33), we obtain

$$\tau \mathbf{v} \nabla f(\mathbf{r}, \varepsilon(\mathbf{v}), t) + f(\mathbf{r}, \varepsilon(\mathbf{v}), t + \tau) = f_0 \tag{2.1.34}$$

Then we multiply the expression (2.1.34) by $\varepsilon D(\varepsilon) \mathbf{v}$ on both sides and integrate over all possible energies using the relation $\int_{\varepsilon} f_0 \varepsilon D(\varepsilon) \mathbf{v} d\varepsilon = 0$ [57]:

$$\int_{\varepsilon} \tau \mathbf{v} \cdot \nabla f(\mathbf{r}, \varepsilon(\mathbf{v}), t) \mathbf{v} \varepsilon D(\varepsilon) d\varepsilon + \mathbf{q}(\mathbf{r}, \mathbf{v}, t + \tau) = 0 \tag{2.1.35}$$

Assuming that the relaxation time τ does not depend on the system's energy and the system has reached a quasi-equilibrium state, then equation (2.1.34) becomes $\nabla f = \frac{df_0}{dT} \nabla T$ [57] and

$$\mathbf{q}(\mathbf{r}, t + \tau) = -\mathbf{k} \nabla T(\mathbf{r}, t) \quad (2.1.36)$$

where $\mathbf{k}$ is the conductivity tensor and

$$\mathbf{k} = \int \tau \mathbf{v} \mathbf{v} \frac{df_0}{dT} \varepsilon D(\varepsilon) d\varepsilon \quad (2.1.37)$$

For isotropic materials $\mathbf{k} = k\mathbf{I}$, where $\mathbf{I}$ is the unit matrix and k is a constant. Hence, equation (2.1.36) reduces to the SPL model. However, in this derivation it was assumed that the considered non-equilibrium state is not far away from equilibrium.

It is important to notice that the classical Boltzmann equation, which does not contain fluctuation terms, is not a closed one, and there is no rigorous solution to the closure problem for the system of moment equations in the theory of turbulence based on hydrodynamic equations derived from the Boltzmann equation.

In 1987, Alekseev [58] derived a generalised Boltzmann equation

$$\frac{\partial \hat{f}}{\partial t} + \mathbf{v} \cdot \frac{\partial \hat{f}}{\partial \mathbf{r}} + \frac{\mathbf{F}}{m} \cdot \frac{\partial \hat{f}}{\partial \mathbf{v}} = J(f) \quad (2.1.38)$$

where $J(f)$ is the collision term and

$$\hat{f} = f - \tau \left[\frac{\partial f}{\partial t} + \mathbf{v} \cdot \frac{\partial f}{\partial \mathbf{r}} + \frac{\mathbf{F}}{m} \cdot \frac{\partial f}{\partial \mathbf{v}} \right] \quad (2.1.39)$$

but equation (2.1.38) not valid in general because it describes the case of gases only.

2.1.10 Dual-Lag Model

The dual-lag model was proposed by Tzou [59], where he introduced two time lags: the heat flux time lag τ_q and the temperature gradient time lag τ_T :

$$q''(x, t + \tau_q) = -k \frac{\partial T}{\partial x}(x, t + \tau_T) \quad (2.1.40)$$

where $-\infty < x < +\infty, 0 < t < +\infty$ and k denotes thermal conductivity of the material.

This model represents a new type of constitutive relation between the heat flux and the temperature gradient, which supercedes the Fourier law at small scales.

The relation (2.1.40) results in the new form of energy conservation equation, the general form of which is

$$\frac{\partial T}{\partial t}(x, t) = -\frac{1}{\rho C_p} \frac{\partial q(x, t)}{\partial x}; \quad -\infty < x < +\infty, \quad 0 < t < +\infty \quad (2.1.41)$$

The corresponding heat transport equation is [60]

$$\frac{\tau_q}{\alpha} \frac{\partial^2 T}{\partial t^2} + \frac{1}{\alpha} \frac{\partial T}{\partial t} = \frac{\partial^2 T}{\partial x^2} + \tau_T \frac{\partial^3 T}{\partial x^2 \partial t} + o(\tau_q) + o(\tau_T); \quad (2.1.42)$$

$$-\infty < x < +\infty, \quad 0 < t < +\infty$$

Equation (2.1.42) shows that the lagging behaviour should be taken into account for the processes whose characteristic time scales are comparable to τ_q and τ_T .

The general solution of equation (2.1.42) is not obtained yet, though the results of the numerical simulation agree quite satisfactory with most of the experimental results [59, 61, 62].

It was shown by Kulish and Novozhilov [63, 64] that the dual-lag model is equivalent to the single-lag model if $\tau_q = 0$. However, this model is still phenomenological, though it describes experimental data well.

2.1.11 Langevin Equation

Some attempts to describe the processes happening in heat transfer, probably could be done using the Langevin equation

$$m \frac{d^2 \mathbf{r}}{dt^2} = -\frac{k_B T}{D} \frac{d\mathbf{r}}{dt} + \mathbf{F}(t) \quad (2.1.43)$$

where $\mathbf{F}(t)$ is the noise term, D stands for the diffusion coefficient, k_B represents the Boltzmann constant and $\mathbf{r}$ satisfies the following condition:

$$\langle \mathbf{r}^2 \rangle = -6D \left[-\frac{mD}{kT} \left(e^{-\frac{kT}{mD}t} - 1 \right) - t \right]. \quad (2.1.44)$$

This solution describes two regimes:

- a) the ballistic regime, for small t when $\langle \mathbf{r}^2 \rangle \approx \frac{3kT}{m} t^2$,
- b) the diffusion regime, for large t , when $\langle \mathbf{r}^2 \rangle \approx 6Dt$.

However, the Langevin equation cannot be used to obtain the expression for the time lag as it is limited by its very nature: it describes Brownian motion in a potential field which is not suitable for solids.

2.1.12 Summary

It was shown that modern models suffer from many significant imperfections. All this requires development of a new, non-phenomenological model of energy transport. Recent discovery [65] shows that, perhaps, the only basis on which such a model can be developed, is the quantum theory.

2.2 The Quantum Theory Point of View

In 1859, Gustav Kirchhoff provided a theorem on blackbody radiation [66]. A blackbody is an object that absorbs all the energy that falls upon it and, because it reflects no radiation, it would appear completely black to an observer. A blackbody also appears to be a perfect emitter and Kirchhoff proved that the emitted energy, E , depends on the temperature T and the frequency ν of the emitted energy only, i.e.

$$E = J(T, \nu) \tag{2.2.1}$$

He challenged scientists to find the function J .

In 1879, Stefan [67] proposed, using experimental data, that the total energy emitted by a hot body is proportional to the fourth power of the absolute temperature. However, in general, this proposition stated by Stefan is false. The same conclusion was reached in 1884 by Boltzmann [68] for blackbody radiation, this time the attempt was made from the theoretical considerations with the use of thermodynamics and Maxwell's electromagnetic theory. This result, now known as the Stefan-Boltzmann law, does not fully answer Kirchhoff's challenge since it does not answer the question for specific wavelengths.

In 1896, Wien [69] proposed a solution to the Kirchhoff challenge. However, although his solution matches experimental observations closely for small values of the wavelength, it was shown by Rubens et al. [70] that it breaks down in the far infrared.

Heinrich Rubens visited Max Planck in October 1900 and tried to explain to him his results, which he achieved with Ferdinand Kurlbaum for the spectrum of cavity radiation up to the longest waves [71]. Within a few hours of Rubens leaving Planck's house, Planck had guessed the correct formula for the Kirchhoff's J function. This guess fitted experimental evidence at all wavelengths very well, but nevertheless Planck was not satisfied with this and tried to give a theoretical derivation of the formula. To do this, he made the unprecedented step of assuming that the total energy is made up of indistinguishable energy elements - quanta of energy.

Planck himself gave credit to Boltzmann for his statistical method but it should be noted that Planck's approach was fundamentally different. However, theory had now deviated from experiment and was based on a hypothesis with no experimental basis [72, 73]. For this work, Planck won the 1918 Nobel Prize for Physics.

In 1901, Ricci and Levi-Civita published "Absolute differential calculus". It had been Christoffel's discovery of 'covariant differentiation' in 1869 [74], which allowed Ricci to extend the theory of tensor analysis to Riemannian space of n dimensions. The Ricci and Levi-Civita definitions were thought to give the most

general formulation of a tensor. This work was not done with quantum theory in mind but, as so often happens, the mathematics necessary to embody a physical theory had appeared at precisely the right moment.

In 1905, Einstein was busy with the examination of the photoelectric effect [75]. The photoelectric effect is the release of electrons from certain metals or semiconductors by the action of light. The electromagnetic theory of light gives results at odds with experimental evidence. Einstein proposed a quantum theory of light to solve the difficulty and after that he realized that Planck's theory made the implicit use of the light quantum hypothesis. Already by 1906 Einstein had correctly guessed that energy changes occur in a quantum material oscillator in jumps which are multiples of $\hbar\nu$, where $\hbar$ is Planck's reduced constant and ν stands for the wave frequency. Einstein received the Nobel Prize for Physics in 1922 for his work on the photoelectric effect.

In 1913, Niels Bohr wrote a revolutionary paper on the hydrogen atom, as he discovered the major laws of the spectral lines [76]. This work earned Bohr the 1922 Nobel Prize for Physics. Later, Arthur Compton [77] derived relativistic kinematics for the scattering of a photon (a light quantum) off an electron at rest in 1923.

While working in the context of the light-quantum hypothesis [78], Einstein assumed that there should be a component of stimulated emission, as in the classical theory. While it was natural for an incoming wave to stimulate the emission of another wave of the same

frequency, the same could be true of light-quanta only if one assumed quanta of the same frequency to be mutually dependent. The assumption of stimulated emission gave light-quanta a quasi-wave-like behaviour, and the assumption that the radiation should tend to infinity with temperature, ensured that this aspect of the classical theory was wholly retained. The importance of the latter assumption was noticed, apparently, first by Eddington [79].

However, there were concepts in the new quantum theory which still gave major worries to many leading physicists. Einstein, in particular, always worried about the element of 'chance' which had entered physics. In fact, Rutherford had introduced spontaneous effect when discussing radioactive decay in 1900. In 1924, Einstein wrote [80]:

There are therefore now two theories of light, both indispensable, and - as one must admit today despite twenty years of tremendous effort on the part of theoretical physicists - without any logical connection.

In the same year, 1924, Bohr et al. [81] made important theoretical proposals regarding the interaction of light and matter, which rejected the photon. Although the proposals were the wrong way forward, they appeared to be able to stimulate important experimental work. Bohr raised several paradoxes in his work:

- a) How can energy be conserved, when some energy changes are continuous and some are discontinuous, i.e. change by quantum amounts?

b) How does the electron know when to emit the radiation?

Einstein had been greatly puzzled by paradox (b), and Pauli quickly told Bohr that he did not believe his theory. Further experimental works soon proved the existence of electron. Other ways had to be found to resolve these paradoxes.

Up to this stage, quantum theory was set up in Euclidean space and used Cartesian tensors of linear and angular momentum. However, quantum theory was about to face a completely new approach.

The year 1924 saw the publication of another fundamental paper. It was written by Satyendra Nath Bose and was rejected by a referee for publication. Bose then decided to send the manuscript to Einstein who immediately saw the importance of Bose's work and arranged for its publication [82]. Bose proposed different states for the photon. He also proposed that there is no conservation of the number of photons. Instead of statistical independence of particles, Bose put particles into cells and talked about statistical independence of these cells. Time has shown that Bose was right on all these points.

Another work was going on at almost the same time as Bose's, which was also of fundamental importance. The doctoral thesis of Louis de Broglie [83] was presented to the scientific audience where the author extended the particle-wave duality from light to all particles, in particular to electrons.

In 1926, Schrödinger published a paper giving his equation for the hydrogen atom and heralded the birth of wave mechanics. Schrödinger introduced operators associated with each dynamical variable [84, 85].

The year 1926 saw the complete solution of the derivation of Planck's law after 26 years. It was solved by Paul Dirac [86]. Also in the same year Born abandoned the causality of traditional physics.

Heisenberg wrote his first paper on quantum mechanics in 1925 [87], and two years later he stated his uncertainty principle [88]. It postulates that the process of measuring the position x of a particle disturbs the particle's momentum p , so that

$$\Delta x \cdot \Delta p \geq \hbar \quad (2.2.2)$$

$$\hbar = \frac{h}{2\pi} \quad (2.2.3)$$

where Δx is the uncertainty of the position and Δp represents the uncertainty of the momentum. Here h is Planck's constant and $\hbar$ is usually called the 'reduced Planck's constant'

Heisenberg's work used matrix methods, which became possible by the work of Arthur Cayley [89] on matrices 50 years earlier. In fact, 'rival' matrix mechanics derived from Heisenberg's work and wave mechanics resulting from Schrödinger's work now entered the arena.

Also in 1927 Bohr stated that space-time coordinates and causality are complementary [90]. Pauli realised that spin, one of the particle's states proposed by Bose, corresponded to a new kind of tensor, one not covered by the Ricci and Levi-Civita work of 1901. However, the mathematics of this had been anticipated by Élie Cartan, who introduced a 'spinor' as part of a much more general investigation in 1913 [91].

Paul Dirac, in 1928, gave the first solution of the problem of expressing quantum theory in a form, which was invariant under the Lorentz group of transformations of special relativity [92]. He expressed d'Alembert's wave equation (see, for example, [20]) in terms of operator algebra.

It is interesting, that the uncertainty principle had a complicated destiny and was not accepted by everyone. Its most outspoken opponent was Albert Einstein. He devised a challenge to Niels Bohr, which he made at a conference which they both attended in 1930. Einstein suggested an example of a box filled with radiation with a clock fitted in one side. The clock is designed to open a shutter and allow one photon to escape. It is proposed to weigh the box after some time and then the photon energy and its time of

escape can both be measured with arbitrary accuracy. Of course this was not meant to be an actual experiment, only a 'thought experiment'.

Nevertheless, Bohr had the final triumph, for the next day he found a very elegant solution. The mass is measured by hanging a compensation weight under the box. This in turn imparts a momentum to the box and there is an error in measuring the position. Time, according to relativity, is not an absolute value and the error in the position of the box translates into an error in measuring the time.

Although Einstein has never believed that the uncertainty principle is true, he accepted it after Bohr's explanation.

In 1932 von Neumann put quantum theory on a firm theoretical basis [93]. Some of the earlier works had lacked mathematical rigour, but von Neumann put the whole theory into the setting of operator algebra and the era of quantum mechanics has begun.

2.3 Superfluidity

2.3.1 Brief History Overview

The pioneering observation of the phenomenon known as “superfluidity” was made in liquid 4-He in 1938 simultaneously by Kapitza in Moscow [94] and Allen and Misener in Cambridge [95]. It was known previously, that liquid helium did not freeze under its own pressure down to the lowest attainable temperatures, and during the 1930s, scientists knew that something unusual is happening below the characteristic temperature ($\sim 2.17\text{ K}$), which is known as the “lambda temperature”. The experiments showed that below the “lambda point” the value of heat flow is not proportional to the temperature gradient and the liquid flows so easily as if the very phenomenon of viscosity did not exist at all. Kapitza called this abnormal behaviour of the liquid “superfluidity”.

A few months later after the experiments, Fritz London gave an explanation to the phenomena observed: as the He atom possesses even number of elementary particles (2 protons, 2 neutrons, 2 electrons), then the many-body wave function of the system should be symmetric under the exchange of any two atoms, i.e. obey the Bose statistics [96]. Earlier, Einstein had studied the behaviour of the gas of non-interacting particles and shown that, below some characteristic temperature, it should have some peculiar behaviour, now known as Bose-Einstein condensation (BEC) [97]. When Einstein made this suggestion, it was believed that this behaviour was due to the model of a non-

interacting gas and was suspected to disappear as soon as the interatomic interactions were taken into account.

After the suggestion of London [96], Laszlo Tisza proposed the idea of explaining this phenomenon by the “two-fluid” model [24]. This suggests that the “condensate” behaves like a fluid completely without friction and the rest behaves like normal one. The prediction he made from this theory was about the collective excitation where the two aforementioned components oscillate out of phase.

Later, Landau suggested the quantitative way of “two-fluid” description of liquid He-II (because of wartime he seemed to be unaware of Tisza’s work) and never proposed there the very idea of BEC [25]. He proposed the first introduction into the condensed-matter physics, such that at sufficiently low temperatures, the total energy, momentum, etc. of the system can be regarded as a sum of those carried by the quasiparticles. The quasiparticles, identified by Landau, are of two types: phonons (quantized sound waves with the energy ε , where $\varepsilon = cp$, c – speed of sound and p is the momentum) and “rotons”, which he has regarded as corresponding to quantized rotational motion and to which he has assigned energy spectrum. The main prediction of this theory is that at sufficiently low temperatures, the contribution of rotons to the specific heat is negligible, which is only due to the phonons at these temperatures and proportional to T^3 .

For the development of the theory of flow properties of He-II, Landau assumed the existence of two components: the “superfluid” component, which is associated with the part of the liquid that remains in its ground state, and a “normal” component, which

corresponds to the quasiparticles. The superfluid component is said to have zero entropy and flowing irrotationally (for the superfluid velocity $\mathbf{v}_s$: $\nabla \times \mathbf{v}_s = 0$), but the normal component behaves like the usual viscous liquid. From these postulates, Landau was able to make three significant predictions:

- a) if the liquid flows relative to the vessel containing it at a velocity smaller than some velocity v_c , now known as the critical Landau velocity, then it is possible that it does so without dissipation, or the flow would be unstable for the creation of the quasiparticles.
- b) if the boundaries are slowly rotating, then only some fraction of the liquid, which corresponds to the normal component, rotates with them, and Landau gave an expression for that fraction in terms of the excitation spectrum.
- c) it is possible to create an oscillation regime (now known as second sound), in which the normal and superfluid components oscillate out of phase.

The last two predictions were verified in the Soviet Union several years after the publication, but the first one appeared to be hard to prove: it was only recently that Landau's critical velocity has been measured.

Although the Landau's two-fluid model of superfluidity explains the basics of the phenomenon in question, its approach is still phenomenological, i.e. the main assumptions are made in the intuitive way but not through explicit demonstration (for example, using Bose statistics). This gap was partially filled by Bogoliubov in 1946 [98], by his proposition of “many-body problem”. The dilute gas of atoms, obeying the Bose statistics and interacting by interatomic interaction, was described. The assumption that was made is based on the idea that such a system, like a completely free Bose gas, would manifest the phenomenon of BEC and the energy spectrum for large momentum p corresponds approximately to the simple excitation of free atoms from the condensate ($\epsilon(p) = \frac{p^2}{2m}$). However, for smaller values of momenta, it has phonon-like form, postulated by Landau: $\epsilon = cp$. It is interesting, that in this work Bogoliubov never introduced Landau's idea of “rotons”.

Though the Bogoliubov's theory was promising, it could not deal with the real problem of liquid He-II (where the atoms are packed too closely). The first to succeed were Feynman and Cohen (only in 1956) using the earlier Feynman's work [99]. It predicted that the excitation spectrum of the real liquid He-II should go over from the “phonon-like” behaviour $\epsilon(p) = cp$ at small values of momenta (predicted by Bogoliubov) and at larger values - to the “roton-like” form:

$$\varepsilon(p) = \Delta + \frac{(p - p_0)^2}{2m} \quad (2.3.1)$$

where Δ is energy of the normal state of the fluid. Current understanding of the problem is, in some way, a mixture of ideas by London, on one hand [100, 101], and by Landau - on the other. It should be pointed out that these ideas have been developed taking into account the phenomenon of superconductivity, resulting in the point of view that superconductivity is nothing but superfluidity occurring in a charged media, or vice-versa.

The general idea that lies under the modern superfluidity theory in a simple Bose system, like liquid 4-He, is that the superfluid phase is characterised by something that may be called “generalised BEC”. The assumption as follows: at any given time t , it is possible to find a complete orthonormal basis (which actually may depend on time) of single-particle states such that one and only one is occupied by a finite fraction of all the particles, while the number of particles in any other single-particle state is of the order of unity or less. The corresponding wave-function of a single particle $\psi_0(\mathbf{r}, t)$ is called “condensate wave function”, and number N_0 of particles occupying it, is “condensate”. However, it is not necessary that number N_0 is equal to the whole number N of particles in the system, even at zero temperature.

In this sense, the conceptual basis for superfluidity is the following: $\psi_0(r, t) = |\psi_0(r, t)|e^{i\phi(r, t)}$, then the superfluid velocity is

$$v_s(r, t) = \frac{\hbar}{m} \nabla \phi(r, t) \quad (2.3.2)$$

This immediately gives us the irrotational superfluid flow: $\nabla \times v_s = 0$. Then the entropy must be entirely carried by the “normal” component, i.e. the particles occupying the single-particle states other than ψ_0 . These two arguments provide sufficient basis for Landau’s phenomenological two-fluid hydrodynamics. But nevertheless, it should be noted that the “superfluid density” ρ_s , in general, is not equal to N_0/V , where N_0 is the number of particles condensed into ψ_0 ; on the contrary, it is believed, that in the case of liquid 4-He, as $T \rightarrow 0$, ρ_s tends to the total density N/V , while N_0 remains only about 10% of N [102].

If we apply the Stokes theorem in a simply connected region of space, in which $|\psi_0|$ is everywhere nonzero, to the curl of superfluid velocity, then it appears that the integral of v_s around any closed curve is zero.

Another interesting application of the definition of the superfluid velocity is in the case of a region, infinite in one dimension, on which $|\psi_0(r, t)|$ vanishes. This may happen due to several reasons: the liquid can be physically excluded from this region or, while atoms

are present in this region, the particular single-particle state, in which BEC has taken place, happens to have a nodal line there. In both of these cases, we can consider an integral of equation (2.2.1) around a circuit that encloses the one-dimensional region in question, we are no longer allowed to use the Stokes' theorem. This leads to the Onsager-Feynman quantization condition [103]:

$$\oint v_s \cdot dl = \frac{nh}{m}. \quad (2.3.3)$$

This equation can be satisfied by a “vortex”, the pattern of flow, in which is $v_s \sim 1/r$, where r is the perpendicular distance from the “core”. The singularity should appear at this core but this is resolved by the assumption that $|\psi_0|$ vanishes there and so v_s is not defined. In this case the circulation can take any value and vortices tend to be stable only under nonequilibrium conditions. However, in the superfluid systems, the circulation is quantized according to the Onsager-Feynman quantization condition, and the vortices can be metastable even under equilibrium conditions, for sufficiently large times [102].

The most interesting application of this equation occurs in another type of geometry, for example, for the annular region between two concentric cylinders. The radius of the annulus is R and its thickness is d , the ratio d/R is sufficiently small. The superfluid velocity $v_s(r, t)$ is hard to be observed directly, so the value of interest is the mass

current $J(r, t)$. According to Landau and Leggett, in equilibrium, this value is given by the following expression

$$J(r, t) = \rho_s v_s(r, t) + \rho_n v_n(r, t) \quad (2.3.4)$$

where the “superfluid” and “normal” densities ρ_s and $\rho_n = \rho - \rho_s$ are functions of temperature only. “Normal” velocity $v_n(r, t)$ is supposed to behave like velocity of the normal, but not superfluid, liquid.

In two different thought-experiments the cylinders are rotated synchronously with angular velocity ω . This corresponds to the case for quantization condition when $n = 1$: $\omega_c = \hbar/mR^2$. The first experiment starts with the liquid that has temperature above the lambda-point T_λ . The cylinders begin to rotate with the small angular velocity ω , and the observer waits for the thermal equilibrium to be achieved. While $T > T_\lambda$, liquid helium behaves like any other, “normal”, liquid. In the rotating liquid the fluid velocity is simply ωR and the total angular momentum is $I_{cl}\omega$, where the classical moment of inertia is NmR^2 .

Continuing to rotate the cylinders, we start to cool the liquid through the temperature T_λ . Below the lambda-point, the “superfluid” component of the liquid appears and moves with the velocity v_s . This v_s is constrained with the quantization condition and cannot be taken equal to ωR , so it no longer contributes to the circulating current. But still the

quantity v_n is given by the ωR , and the total angular momentum is reduced by $\rho_n(T)/\rho$. Hence, by changing the temperature below or above T_λ , the angular momentum can be reversibly increased or decreased; for $T \rightarrow 0$ it tends to zero, even though the vessel is still rotating.

At larger values of ω ($\omega > \omega_c/2$), the superfluid component will contribute into the angular momentum by the amount $\sim n\omega_c$, where n is the nearest integer to ω/ω_c . That is why, for example, for some values of ω , the velocity of the liquid can exceed that of the container. This remarkable effect (analog of the Meissner effect in superconductors) was first predicted by London [100, 101] and observed by Hess and Fairbank in 1967 [104].

The second thought-experiment may seem close to the first one, but it is conceptually different. We again start with the temperature T_λ , but the liquid rotates much faster than in the previous case: $\omega \gg \omega_c$, so the velocity would be $v = \omega R$. Then we start cooling the system until we reach the temperature T_λ . According to the description above, the superfluid component will take the quantized value of circulation, which makes n closest to ω/ω_c , but ω/ω_c is very large, so the angular momentum is $I_{cl}\omega$. Then, still keeping the temperature on the level of T_λ , we stop the rotation of the cylinders. The contribution of the normal component to the mass current rapidly relaxes to zero, but the superfluid contribution remains and it can be increased or decreased by changing the temperature up and down, without allowing it to exceed the lambda-point T_λ . It means that the system keeps the value of the superfluid motion, even though it is not the equilibrium one. This is called the phenomenon of metastable superflow.

The explanation of this phenomenon requires consideration of interatomic interactions, unlike the Hess-Fairbank effect that can be understood in terms of behaviour of a single atom under the same conditions. It is believed that a noninteracting Bose gas, though in the BEC state, would not behave like this.

The theory, based on the given equations, was proved experimentally. Hence, to describe the main phenomena of superfluidity of 4-He , it can be used not only qualitatively but also quantitatively.

However, in the past 70 years since London's investigations, it is still too hard to verify the existence of Bose-Einstein condensation phenomenon directly. The most information comes from high-energy neutrons scattering and the spectrum of atoms evaporated from the surface of the liquid and only while they both are consistent with the existence of the condensate fraction of approximately 10%.

All said refers to the best known "superfluid" 4-He below the lambda-temperature. But in 1972 it was discovered by Osheroff, Lee and Richardson [105] that the light isotope of helium, 3-He , has not one, but three anomalous phases, at much lower temperature of 3 mK. These phases are referred as "superfluid 3-He ". In this case, the 3-He atom obeys the Fermi statistics, so the superfluidity mechanism for this liquid cannot be as simple as BEC in the case of 4-He . It is believed that fermions form the so-called "Cooper pairs" – some sort of giant diatomic quasimolecules whose characteristic radius is much larger than the usual interatomic distance [106]. These quasimolecules obey the Bose statistics

and hence can undergo BEC. Nevertheless, it should be noted, that the formation of the Cooper pairs and the process of BEC itself are not two independent phenomena: they are deeply connected with each other.

2.3.2 The Phenomenon of BEC

To explain what BEC is, we consider a system of N identical spinless bosons characterised by spatial coordinates $\mathbf{r}_i$ ($i = 1, 2, \dots, N$) with arbitrary interparticle interactions and subject to some external potential (it can be time-dependent), which is assumed to confine the particles in some finite region of space. The system is not necessarily at thermal equilibrium or at a steady state. The many-body wave-functions (wave functions describing interactions between particles) $\psi_N(\mathbf{r}_1 \mathbf{r}_2 \dots \mathbf{r}_N, t)$ must be symmetric with the respect to a change of coordinate of any two particles: $\mathbf{r}_i \rightleftharpoons \mathbf{r}_j$.

The one-particle reduced density matrix can be defined in a standard way for any moment of time t : $\rho(\mathbf{r}, \mathbf{r}', t)$. This matrix can be diagonalised with real eigenvalues [107]. Hence, it is always possible to find a complete orthonormal basis, in general, time independent, of single-particle eigenfunctions $\psi_i(\mathbf{r}, t)$, such that

$$\rho(\mathbf{r}, \mathbf{r}', t) = \sum_i n_i(t) \psi_i^*(\mathbf{r}, t) \psi_i(\mathbf{r}', t) \quad (2.3.5)$$

To define BEC, we say that at any given time t , the system is in the state of BEC if one or more of the eigenvalues $n_i(t)$ is of the order of total number of particles N ; and it gives simple BEC, if one and only one eigenvalue is of the order of N , all others are of the order one.

In case of simple BEC, we shall call the single-particle state $\psi_0(\mathbf{r}, t)$ the condensate wave function and the eigenvalue $N_0(t)$ - the number of particles in the condensate. The superfluid velocity has been already introduced. The more basic definition of BEC includes in itself a hyperfine degree of freedom: the particle is characterised not just by its spatial coordinate $\mathbf{r}_i$, but also by a discrete hyperfine index α_i , so ψ_N is the function of $\{\mathbf{r}_i, \alpha_i\}, i = 1, 2 \dots N$. Thus, in general, the density matrix becomes

$$\rho(\mathbf{r}\alpha, \mathbf{r}'\alpha', t) = \sum_i n_i(t) \psi_i^*(\mathbf{r}, \alpha, t) \psi_i(\mathbf{r}', \alpha', t) \quad (2.3.6)$$

The only difference in definitions is that the condensate function $\psi_0(\mathbf{r}, \alpha, t)$ now has a discrete (hyperfine) argument α as well as the continuous $\mathbf{r}$.

In reality, the rigorous results for an interacting Bose system in more than one dimension were obtained, but still they are few and far between [108]. Here are some of them:

- a) For a three-dimensional system in a free space, it was showed [109] that if perturbation theory starting from a noninteracting Bose gas converges, then, at $T = 0$, the system manifests BEC. But still this fact shows nothing about the condensed fraction $N_0(0)/N$ and the critical temperature T_c .
- b) In practice, a continuous medium is replaced by a lattice gas and the interatomic interactions are modeled by a hardcore on-site repulsion, and, for the case of half-filling, the existence of BEC at $T = 0$ has been proved [81].
- c) The theorem concerning the (non)occurrence of BEC in a K -dimensional space proves that for $K \leq 2$ BEC cannot occur at any finite temperature [110].
- d) In the case of $K = 3$, there is no upper limit for T_c , but there is one for $N_0(T)/N$, which is completely independent of the nature and the sign of the interatomic potential [111].
- e) For a Bose gas with an interaction that is everywhere repulsive and has a space integral V_0 , it is possible to place a stronger upper limit on the condensate for small V_0 [112].

2.3.3 The Effective Interaction in a Cold Dilute Gas

For consideration of atom-atom interactions in a gas at very low temperatures and densities at the BEC conditions, we assume that two colliding atoms can be “tagged” (as

this will allow us not to take into account the effect of indistinguishability of the atoms) and neglect the aforementioned hyperfine degree of freedom to assume that for every atom its initial and final values in a collision are equal [113, 114]. Then the only relevant variable for a pair of atoms is then their relative coordinate $\mathbf{r}$ and the outcome of the collision is determined by the value of kinetic energy

$$E \equiv \frac{\hbar^2 k_w^2}{2m_r} \quad (2.3.7)$$

where m_r is the reduced mass (m_r is defined as $m_r = \frac{m_1 m_2}{m_1 + m_2}$, where m_1 and m_2 are masses of these two atoms) and k_w is the relative wave factor.

If we consider the true interatomic potential $V_{\text{at}}(r)$ then at short distances it may not even be definable, but at short distances of about $\gtrsim 5 \text{ \AA}$, $V_{\text{at}}(r)$ should be well-defined and well-approximated by the lowest-order van der Waals interaction C_6/r^6 (here C_6 is a constant, different for different atoms). This form of potential defines a characteristic van der Waals length, i.e.

$$r_0 \equiv \left(\frac{2m_r C_6}{\hbar^2} \right)^{\frac{1}{4}} \quad (2.3.8)$$

the physical meaning of which is the typical extent of the last bound state in the potential. The order of this length is $50 \text{ \AA}$, which is much larger than the typical molecular dimension. The associated characteristic energy

$$E_c \sim \frac{\hbar^2}{m_r r_0^2} \quad (2.3.9)$$

is of the order 0.1-1 mK.

It is important to note that for all the alkalis (including hydrogen) the values of the thermal energy $k_B T$ characterising BEC conditions are small compared to the above energy. It appears that for $l \neq 0$, the probability of finding two atoms at a distance r_0 one from another at a collision falls off as $(k_w r_0)^{2l}$, where l is the relative angular momentum, the effective scattering amplitude is negligible comparing to one arising from the s-wave scattering ($l=0$), so we can use the s-wave case only [115].

The Schrödinger equation solution for this case is

$$\psi(r) = C \frac{\sin[k(r - a_s)]}{r} \quad (2.3.10)$$

where a_s is the (zero-energy) s-wave scattering length and, depending on the value of the potential, may have different sign [116] and C is an arbitrary constant.

The s-wave scattering length is, in general, a function of chemical and isotopic species involved and the hyperfine indices of the two atoms, even of the magnetic and/or laser field. It can be obtained experimentally by the technique of photoassociative spectroscopy [117] or from the knowledge of the atomic mean free path in the gas [118-121].

2.3.4 Recent Discoveries

As was mentioned before, second sound was first successfully measured in 1966 [34]. However, not only second sound appears to exist in superfluids. The existence of third, fourth and even fifth sound was predicted theoretically, and then discovered experimentally in superfluid He [122-125].

Bose-Einstein condensate was created from rubidium atoms in 1995 [126]. After this, the prospect of creating a similar sort of condensate made from fermionic atoms naturally arose. This substance would form a superfluid by the BCS mechanism. The way to create a Fermionic condensate (superfluid phase, which is formed by fermionic particles at low temperatures) from pairs of ultra-cold fermionic atoms has been recently found out involving potassium atoms [127]. Under certain conditions, these fermion pairs can form diatomic molecules which are able to undergo BEC and behave like superfluid. On the other hand, the fermions (most notably superconducting electrons) form

aforementioned Cooper pairs which also show the state of superfluidity. This recent work with ultra-cold atomic gases has made possible for scientists to explore the region in between these two extremes, known as the BEC-BCS crossover.

Surprisingly, even such thing as supersolids may also have been discovered in 2004 by physicists at Pennsylvania State University. When 4-He is cooled below about 200 mK under sufficiently high pressures, some fraction (about 1%) of the solid appears to become supersolid [128-131]. It was shown that by quench cooling or lengthening the annealing time, thus increasing or decreasing the defect density respectively, via torsional oscillator experiment, the supersolid fraction could be made to range from 20% to completely non-existent. It may be supposed that the supersolid nature of 4-He is not intrinsic to 4-He but it appears to be a property of 4-He and disorder [132, 133]. Some emerging theories proposed that the supersolid signal observed in 4-He was actually an observation of either a superglass state [134] or intrinsically superfluid grain boundaries in the 4-He crystal [135].

However, by now superfluidity is still in the extreme lack of theoretical explanation. This outstanding physical phenomenon requires further and deeper investigation of the very basic processes that cause all these remarkable properties of the matter.

Chapter 3. Theoretical Part

3.1 Implications from the Schrödinger Equation

Quantum mechanics is deeply connected with the macroscopic phenomena, though it is not obvious from a first look. The Schrödinger equation describes one of the most basic levels of the world – the behaviour of the elementary particles. However, it was shown by Landau [115] that several interesting equations follow from this basic expression:

$$i\hbar \frac{\partial \psi}{\partial t} = -\frac{\hbar^2}{2m} \nabla^2 \psi + U(x, y, z) \psi \quad (3.1.1)$$

where ψ is the wave function of the particle, $\hbar$ stands for the reduced Planck's constant, U represents the potential energy of the particle in the external field and m denotes the mass of the particle.

Let us use the following expression for the wave function:

$$\psi = ae^{-\frac{i}{\hbar}\theta} \quad (3.1.2)$$

Upon applying equation (3.1.2) to the Schrödinger equation and performing differentiation, we obtain

$$a \frac{\partial \theta}{\partial t} - i\hbar \frac{\partial a}{\partial t} + \frac{a}{2m} (\nabla \theta)^2 - \frac{i\hbar}{2m} a \nabla^2 \theta - \frac{i\hbar}{m} \nabla \theta \cdot \nabla a - \frac{\hbar^2}{2m} \nabla^2 a + Ua = 0 \quad (3.1.3)$$

By definition, a and θ are real, but in this equation there are purely real and imaginary terms. For equation (3.1.3) to be true, each part of it (both real and imaginary) should be equal to zero:

$$\frac{\partial \theta}{\partial t} + \frac{1}{2m} (\nabla \theta)^2 + U - \frac{\hbar^2}{2m} \nabla^2 a = 0 \quad (3.1.4)$$

$$\frac{\partial a}{\partial t} + \frac{a}{2m} \nabla^2 \theta + \frac{1}{m} \nabla \theta \cdot \nabla a = 0 \quad (3.1.5)$$

Neglecting the term with $\hbar^2$ in the first equation due to its smallness, we obtain

$$\frac{\partial \theta}{\partial t} + \frac{1}{2m} (\nabla \theta)^2 + U = 0 \quad (3.1.6)$$

which is simply the classical Hamilton-Jacobi equation for the action θ of the particle and which will play an important role in our investigations in the theoretical part of this study. It is obvious, that, as $\hbar \rightarrow 0$, classical mechanics is valid as long as quantities of the first (and not only the zero) order in $\hbar$ are included.

Multiplying equation (3.1.5) by $2a$, we get

$$\frac{\partial a^2}{\partial t} + \operatorname{div} \left(a^2 \frac{\nabla \theta}{m} \right) = 0 \quad (3.1.7)$$

The meaning of this equation is rather obvious: a^2 is the probability density of finding the particle at some point in space (as $|\psi|^2 = a^2$). If we set

$$\frac{\nabla \theta}{m} = \frac{\mathbf{p}}{m} \quad (3.1.8)$$

the term $\frac{\mathbf{p}}{m}$ would be the classical velocity $\mathbf{v}$ of the particle. It means that the above equation is simply the equation of continuity, which shows that the probability density

“moves” according to the laws of classical mechanics with the classical velocity $\mathbf{v}$ at every point.

3.2 Implications to the Navier-Stokes and Diffusion Equation

In this section we show that the existence of a narrow wave packet leads to the flow becoming inviscid.

Let us consider the Navier-Stokes equation written for a case of constant density and viscosity fluid [17]:

$$\rho \left[\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v} \cdot \nabla) \mathbf{v} \right] = -\nabla p + \mu \nabla^2 \mathbf{v} + \left(\lambda + \frac{\mu}{3} \right) \nabla (\nabla \cdot \mathbf{v}) \quad (3.2.1)$$

where ρ , λ and μ are density, second viscosity, and dynamic viscosity of the fluid respectively, p is the pressure and $\mathbf{v}$ denotes the velocity.

Under the assumption of the fluid being incompressible, we can re-write the Navier-Stokes equation in a much simpler form, since $\nabla \cdot \mathbf{v} = \mathbf{0}$:

$$\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v} \cdot \nabla) \mathbf{v} = -\frac{1}{\rho} \nabla p + \nu_k \nabla^2 \mathbf{v} \quad (3.2.2)$$

where $\nu_k = \mu/\rho$ denotes kinematic viscosity.

Since the velocity at a certain location x is proportional to the gradient of the corresponding de Broglie's wave's phase at the same location [17], i.e.,

$$\mathbf{v}(x) = \nabla \tilde{\theta} = \frac{\hbar}{m} \nabla \theta = -\frac{i\hbar}{m} \frac{\nabla \psi}{\psi} \quad (3.2.3)$$

where $\frac{\hbar}{m} \theta = \tilde{\theta}$ is the velocity potential, we can re-write the Navier-Stokes equation again, but now in terms of the phase of the de Broglie's wave corresponding to the moving fluid particle, taking into account that

$$(\mathbf{v} \cdot \nabla) \mathbf{v} = (\nabla \tilde{\theta} \cdot \nabla) \nabla \tilde{\theta} = \frac{1}{2} \nabla (\nabla \tilde{\theta} \cdot \nabla \tilde{\theta}) - \nabla \tilde{\theta} \times (\nabla \times \nabla \tilde{\theta}) = \frac{1}{2} \nabla (\nabla \tilde{\theta} \cdot \nabla \tilde{\theta}) \quad (3.2.4)$$

and

$$\nabla^2 \mathbf{v} = \nabla(\nabla \cdot \mathbf{v}) - \nabla \times (\nabla \times \mathbf{v}) = \nabla(\nabla \cdot \nabla \tilde{\theta}) - \nabla \times (\nabla \times \nabla \tilde{\theta}) = \nabla(\nabla^2 \tilde{\theta}) \quad (3.2.5)$$

since $\nabla \times \nabla \equiv 0$; therefore

$$\nabla \left[\frac{\partial \tilde{\theta}}{\partial t} + \frac{1}{2} (\nabla \tilde{\theta} \cdot \nabla \tilde{\theta}) \right] = \nabla \left[-\frac{p}{\rho} + \nu_k \nabla^2 \tilde{\theta} \right] \quad (3.2.6)$$

or

$$\frac{\partial \tilde{\theta}}{\partial t} + \frac{1}{2} (\nabla \tilde{\theta} \cdot \nabla \tilde{\theta}) = -\frac{\Delta p}{\rho} + \nu_k \nabla^2 \tilde{\theta} \quad (3.2.7)$$

where Δp is the difference between the actual pressure p and a certain reference pressure p_0 .

Now, let us substitute the following expression for $\tilde{\theta}$:

$$\tilde{\theta} = -2\nu_k \ln \tilde{\psi} \quad (3.2.8)$$

then the Navier-Stokes equation becomes:

$$\frac{\partial \tilde{\psi}}{\partial t} - \nu_k \nabla^2 \tilde{\psi} = \frac{\Delta p}{2\mu} \tilde{\psi} \quad (3.2.9)$$

which is the reaction-diffusion equation written in terms of the scalar function $\tilde{\psi}$.

From equation (3.2.3), it follows that

$$\tilde{\theta} = -\frac{i\hbar}{m} \ln \psi \quad (3.2.10)$$

But on the other hand, here we used the transformation $\tilde{\theta} = -2\nu_k \ln \tilde{\psi}$ for the Navier-Stokes equation. It follows that the relation between $\tilde{\psi}$ and ψ is

$$\tilde{\psi} = \psi \exp\left(\frac{i\hbar}{2m\nu_k}\right) \quad (3.2.11)$$

Both $\tilde{\psi}$ and ψ represent the same state of the particle as their amplitudes are equal and, therefore, the probability densities determined by them are the same. Substituting the expression (3.2.11) into (3.2.9), we obtain:

$$\frac{\partial \psi}{\partial t} - v_k \nabla^2 \psi = \frac{\Delta p}{2\mu} \psi \quad (3.2.12)$$

which is the reaction-diffusion equation for the wave function corresponding to the de Broglie's wave of a fluid particle moving with velocity $\mathbf{v}$.

From equation (3.2.3) it follows that in this case $\nabla \times \mathbf{v} = 0$ as the velocity potential exists. It means that the fluid becomes inviscid, i.e., superfluid! But this superfluid regime exists as long as wave packet exists only. The goal of the next section is to find the wave packet life time span.

3.3 The Fundamental Time Lag

As we already saw, there were many efforts to prove the existence and to derive a general expression for the time lag phenomenon, but they were all unsuccessful because the principles that were used for these derivations are not suitable for all media.

In my research, I used the most basic theory for the investigations in this area – quantum theory, as its laws are suitable for all materials and all media in nature. The main assumption used in this part is that a narrow wave packet forms as an immediate result of media excitation. In other words, in an initial moment of excitation a medium behaves like a narrow wave packet which breaks down due to its widening after some time interval τ .

The main assumption of the model developed in this chapter is that narrow wave packets of the excited particles are formed in the media immediately upon absorbing an external stimulus. These wave packets remain narrow during a certain time span τ , during which flow behaves as inviscid (superfluid).

We now provide an estimate of the life span of a narrow wave packet. In other words, we wish to estimate how long the heat wave (second sound) will exist in the transporting medium using the principles of quantum theory.

We use the non-relativistic Schrödinger equation

$$i\hbar \frac{\partial \psi}{\partial t} = -\frac{\hbar^2}{2m} \nabla^2 \psi + U(x, y, z) \psi \quad (3.3.1)$$

which is applicable to all states of aggregation of matter, including solids. Here $U(x, y, z)$ is the potential energy of the particle in the applied field, ψ denotes the corresponding wave function, $\hbar$ represents the reduced Planck's constant and m stands for the non-relativistic mass of the particle.

From the basic principles of quantum theory, it follows that any freely moving particle can be associated with the flat wave described by a wave function, implying it has energy E and momentum $\mathbf{p}$:

$$\psi(\mathbf{r}, t) = A \exp[-i(\omega t - \mathbf{k} \cdot \mathbf{r})] \quad (3.3.2)$$

where $\mathbf{r}$ is the radius-vector of an arbitrary point in space, A represents the wave amplitude and t denotes time [83]. As was mentioned before, the frequency ω of the wave and its wave number $\mathbf{k}$ are related to the particle energy and momentum by the following relationships:

$$E = \hbar \omega \quad (3.3.3)$$

$$\mathbf{p} = \mathbf{k}\hbar \quad (3.3.4)$$

These equations are called de Broglie's equations. Using them ($\omega = E/\hbar$, $\mathbf{k} = \mathbf{p}/\hbar$) and the equation for the flat wave given by equation (3.3.2), we can obtain the following expression:

$$\psi(\mathbf{r}, t) = A \exp \left[-i \left(\frac{Et}{\hbar} - \frac{\mathbf{p}\mathbf{r}}{\hbar} \right) \right] \quad (3.3.5)$$

This kind of wave is called de Broglie's wave and $\psi(\mathbf{r}, t)$ here is the wave function. This function determines the probability of finding the particle in a given point at a given moment of time.

It is obvious that the square of the modulus of the wave function is equal to a constant:

$$|\psi(x, t)|^2 = A^2 \left| e^{-\frac{i\theta}{\hbar}} \right|^2 = A^2 \left| \cos \frac{\theta}{\hbar} + i \sin \frac{\theta}{\hbar} \right|^2 = A^2 \quad (3.3.6)$$

From this we can conclude that the flat wave fills the space with equal density. This is why it is impossible to describe the movement of a particle by a flat monochromatic wave only.

For the wave description of a particle, it is necessary to take not a single monochromatic wave but a set of waves that have close frequencies, which is called a wave packet. Hence, the wave packet is a combination of waves with different frequencies describing some formation that possesses wavelike features and travels as a unit and, in general, is bounded in time and space. We can construct such a wave packet, the resulting amplitude of which is appreciably nonzero only in some small region. This region can be regarded as the position of the particle.

Let us form a wave packet from the superposition of the flat waves that have wave numbers in the interval from $k_0 - \frac{\Delta k}{2}$ to $k_0 + \frac{\Delta k}{2}$ (interval $\frac{\Delta k}{2}$ is taken here for a computational convenience). For the sake of simplicity but without loss of generality, we assume that the amplitudes of the waves are constant and equal to $\frac{A}{\Delta k}$ within this interval.

With all these conditions we have the resulting wave function of the packet as

$$\psi(x, t) = \frac{A}{\Delta k} \int_{k_0 - \frac{\Delta k}{2}}^{k_0 + \frac{\Delta k}{2}} \exp[-i(\omega t - kr)] dk \quad (3.3.7)$$

Let us now expand the frequency ω into the Taylor series as a function of k in the vicinity of the point $k = k_0$. It follows

$$\omega(k) = \omega_0 + (k - k_0)\omega'_0 + \frac{(k - k_0)^2}{2}\omega''_0 + \dots \quad (3.3.8)$$

Here $\omega_0 = \omega(k_0)$, $\omega'_0 = \left(\frac{d\omega}{dk}\right)_{k=k_0}$, etc.

Neglecting terms of the order higher than one, we get

$$\omega(k) = \omega_0 + (k - k_0)\omega'_0 + \dots = \omega_0 + \omega_1 + \dots \quad (3.3.9)$$

The truncated term of the second order of magnitude is equal to

$$\omega_2(k) = \frac{(k - k_0)^2}{2} \omega_0'' \quad (3.3.10)$$

Taking into account terms of the first order of magnitude only and integrating the equation (3.3.7) for the wave packet, we obtain the expression for the wave function

$$\psi(x, t) = B e^{-i(\omega_0 t - k_0 x)} \quad (3.3.11)$$

the amplitude of the wave packet B has the form of

$$B = A \operatorname{sinc} \xi, \text{ where } \xi = \frac{\Delta k}{2} (x - \omega_0' t) \text{ and } \operatorname{sinc} \xi = \frac{\sin \xi}{\xi} \quad (3.3.12)$$

Now, let us take into account the neglected influence of the second order terms ω_2 , which are responsible for the phenomenon of dispersion. If the dispersion is zero ($\omega_0'' = 0$), then all the monochromatic waves that form the wave packet are traveling with equal velocities and so the initial form of the wave packet remains the same at all times.

In case when the dispersion is not zero ($\omega_0'' \neq 0$), it appears that the initial configuration of the wave packet begins to change, or, to widen.

Our goal here is to estimate the time of widening of the wave packet. For this purpose, we need to take into account the abovementioned term ω_0 . It is very important, as this term gives us an additional phase in the wave function:

$$\Delta\xi \approx \frac{(\Delta k)^2}{2} \frac{\partial^2 \omega}{\partial k^2} t \quad (3.3.13)$$

which becomes significant when it reaches an order of π (because in the sine function we can neglect the phase only if it is much less than π).

To obtain the moment of time $t = \tau$, when the wave packet begins to widen, we put $\Delta\xi = 2\pi$ in (3.3.13) and get the following expression:

$$\tau \approx \frac{2\pi}{(\Delta k)^2} \frac{1}{\frac{\partial^2 \omega}{\partial k^2}} \quad (3.3.14)$$

Let us see what would happen for de Broglie's waves (Fig.3.1)

Figure 3.1. The form of the wave packet at $t=0$ for de Broglie's waves.

In this case, using the equation for the motion of the de Broglie's wave, the second order term of the Taylor series is:

$$\frac{\partial^2 \omega}{\partial k^2} = \hbar \frac{\partial^2 E}{\partial p^2} \quad (3.3.15)$$

Here we consider the non-relativistic case ($p \ll mc$, where m is the mass of the particle), for which we further obtain $E = \frac{p^2}{m}$

$$\frac{\partial E}{\partial p} = \frac{p}{m}, \quad \frac{\partial^2 E}{\partial p^2} = \frac{1}{m} \quad (3.3.16)$$

Applying these relations to the expression of the estimated time of the widening of the wave packet equation (3.3.14) we have

$$\tau \approx \frac{2\pi\hbar}{(\Delta p)^2} \frac{1}{\frac{\partial^2 E}{\partial p^2}} \approx \frac{m(\Delta x)^2}{h} \quad (3.3.17)$$

where h is Planck's constant.

Using the equation for the traveled distance $\Delta x = c\tau$, where c denotes the group velocity of the wave packet, we can get another form for τ

$$\tau \approx \frac{h}{mc^2} \quad (3.3.18)$$

The physical meaning of this result is as follows: in the beginning of the process, the set of particles of the medium can be regarded as a narrow wave packet, so until it is not yet widened, there will be no diffusion. And the liquid, for example, behaves as superfluid. But after some lapse of time τ , the wave packet loses its wave nature (widens) and diffusive process begins.

This process can be represented by a very simple and intuitive example. Imagine you have a rectangular plaza crowded by people. The person (messenger) in one corner of the plaza needs to deliver a parcel to a person in the diagonal corner of the plaza. If people crowding the plaza are cooperative with each other, then they can transfer the parcel from hand to hand and it will find the addressee quickly. If we look at the plaza from above, we will see the wave corresponding to the process of the parcel transferred from hand to hand. In the other case, when people are not collaborating with the messenger, the latter has to deliver the parcel himself, squeezing his way through the non-cooperative crowd. From above it will look like a diffusive process.

Recall that c denotes the velocity of the particle, which can be associated with the group velocity of the wave packet, which, in turn, is proportional to the gradient of the wave's phase, so in de Broglie's case [17]:

$$c = v(\mathbf{r}) = \frac{\hbar}{m_0} \nabla \theta \quad (3.3.19)$$

where $\frac{\hbar}{m_0} \theta$ is the velocity potential.

This expression matches with the Landau's derivations of macroscopic equations from the Schrödinger equation [115]:

$$\frac{\partial \theta}{\partial t} + \frac{1}{2m} (\nabla \theta)^2 + U = 0 \quad (3.3.20)$$

where U is the potential energy of the particle in the external field, and the whole equation is the Hamilton-Jacobi equation (3.1.6) for the action θ of the particle.

If there is no applied external field, then $U = 0$, and, in the absence of velocity (no phase, no action), there will be no wave and the time of wave packet widening would then be infinite.

However, the finiteness of the phase lag itself implies the existence of the onset of the temperature gradient and the correlating heat flux.

3.4 The Equation for the Second Sound

It was shown by Kulish and Lage [17] that the existence of the narrow wave packets is equivalent to the superfluid state in liquids (see also Leggett [102]) as $\mathbf{v} = \nabla\theta$, or the super-transport state in solids (see Kulish and Chan [15]). However, in their paper Kulish and Lage did not mention this explicitly as well as the phenomena of the wave packet breakdown, where they prove that the flow is irrotational without assuming the liquid to be inviscid. Still, it is not true because the wave packet which they assume to be stable in the course of time does not behave like this and has the feature of widening and hence, the flow remains irrotational only during a short period of time when the wave packet remains narrow.

Much earlier it was shown by Landau (see [136]) that the phenomenon of the second sound is intrinsic to the superfluid state. In other words, a temperature wave exists in the medium

$$\frac{\partial^2 T}{\partial t^2} = u_2^2 \nabla^2 T \quad (3.4.1)$$

where u_2 denotes the velocity of second sound which markedly depends on temperature.

Deriving his equation for second sound, Landau did not use the quantum theory standpoint but the derivations were solely based on the thermodynamics theory.

However, it appears to be possible to derive this expression directly from the equation that describes the motion of a narrow wave packet associated with de Broglie's wave from equation (3.3.15):

$$\frac{\partial^2 \omega}{\partial k^2} = \hbar \frac{\partial^2 E}{\partial p^2} \quad (3.4.2)$$

Here ω is the frequency of the wave, k denotes the wave number, E represents the energy of the wave and p stands for the momentum.

Using the identity for the energy and frequency for wave, obtained by Einstein shown in equation (3.3.3), we arrive at the following equation:

$$\frac{\partial^2 E}{\partial k^2} = \hbar^2 \frac{\partial^2 E}{\partial p^2} \quad (3.4.3)$$

Let us recall the Boltzmann expression for the gas energy

$$E = l_B k_B T \quad (3.4.4)$$

where l_B is some constant (in general, it is proportional to the number of degrees of freedom of the molecules) and k_B denotes the Boltzmann constant. Substituting it into the wave packet's motion equation (3.3.4) and cancelling the constants, we get the following result:

$$\frac{\partial^2 T}{\partial k^2} = \hbar^2 \frac{\partial^2 T}{\partial p^2} \quad (3.4.5)$$

For the momentum, we use equation (3.3.4), which is $\mathbf{p} = \mathbf{k}\hbar$.

At the same time, for the particle of mass m ,

$$p = mu_2 \quad (3.4.6)$$

Combining equations (3.4.5) and (3.4.6), we get the expression for the velocity

$$u_2 = \frac{\hbar k}{m} \quad (3.4.7)$$

It follows then for the velocity and the momentum:

$$u_2^2 = \frac{\hbar k}{p^2}, \quad p^2 = \frac{\hbar k}{u_2^2} \quad (3.4.8)$$

Substituting these expressions into the second sound equation and into the wave motion equation and vice versa, it appears that the equations can be easily transformed one into another:

$$\frac{\partial^2 \omega}{\partial k^2} = \hbar \frac{\partial^2 E}{\partial p^2} \Leftrightarrow \frac{\partial^2 T}{\partial t^2} = u_2^2 \frac{\partial^2 T}{\partial x^2} \quad (3.4.9)$$

It means that the macroscopic equation for the temperature wave (second sound) follows directly from the equation of the wave packet motion, which is based solely on the principles of quantum theory!

Another important conclusion here is that Landau's expression is applicable only for liquids and gases, as derived from the thermodynamics principles only. In this case, however, equation (3.4.9) is applicable for solids as well. That means that the second sound exists in solids!

3.5 The Relation between the Time Lag and Temperature

Let us now analyze equation (3.3.18) from a different point of view. Imagine that we have a test volume of a medium subjected to an external stimulus. As we know, mass is equal to the density of the material ρ times the volume V occupied by the substance:

$$m = \rho V \quad (3.5.1)$$

The volume V can be represented as the cross-section area S of the given volume times its length, which is, in turn, the distance travelled by the single particle during the time τ :

$$V = c\tau \cdot S \quad (3.5.2)$$

As the energy E received by the system equals the cross-section area of the given volume times the energy flux φ , and then we have:

$$S = \frac{E}{\varphi} \quad (3.5.3)$$

Taking into account that $\tau = \frac{V}{cS} = \frac{m\varphi}{\rho cE}$ and $m = \frac{h}{c^2\tau}$, for the wave packet widening time τ we obtain the following expression:

$$\tau = \frac{h\varphi}{c^3\rho\tau E} \quad (3.5.4)$$

or, finally,

$$\tau = \sqrt{\frac{h\varphi}{c^3\rho E}} \quad (3.5.5)$$

Thus, it follows that the time lag depends on the physical qualities of the transporting medium itself (density), velocity of the particle and the ratio $\frac{E}{\varphi}$.

Substituting the Boltzmann law from equation (3.4.4) into the equation (3.5.5) we get

$$\tau = \sqrt{\frac{h\varphi}{c^3\rho l_B k_B T}} \quad (3.5.6)$$

which shows that the time lag is inversely proportional to the square root of the temperature. In general, some other terms here also depend on temperature, but this result is totally in accord with the previous result obtained by Kulish and Chan [15] from statistical physics standpoint.

Chapter 4. Apparent Thermal Diffusivity and Viscosity and Steady State Heat Transfer

4.1 Apparent Thermal Diffusivity and Viscosity

Let us consider the one-dimensional equation describing viscous flow in a semi-infinite domain:

$$\tau_v \frac{\partial^2 u}{\partial t^2} + \frac{\partial u}{\partial t} = \nu_k \frac{\partial^2 u}{\partial y^2}, \quad (y, t) \geq 0 \quad (4.1.1)$$

where τ_v is the relaxation time due to viscous effects, u stands for the medium velocity and ν_k is the kinematic viscosity of the fluid. τ_v and ν_k are assumed to be constant. Equation (4.1.1) is now applied to solve the initial value problem in a semi-infinite domain under the condition of initial equilibrium of the fluid in the domain ($u = u_0$ everywhere at time $t = 0$ and $\frac{\partial u}{\partial t} \Big|_{t=0} = 0$).

We introduce now a new variable $\xi = \frac{y}{\sqrt{\nu_k}}$, the new timelike variable $\vartheta = \frac{t}{\sqrt{\tau_v}}$, and the excess velocity $U_e = u - u_0$. Hence, equation (4.1.1) becomes

$$\frac{\partial^2 U_e}{\partial \vartheta^2} + \sqrt{\tau_v} \frac{\partial U_e}{\partial \vartheta} = \frac{\partial^2 U_e}{\partial \xi^2} \quad (4.1.2)$$

with the initial conditions $U_e = 0$ at $\vartheta = 0$ and $\frac{\partial U_e}{\partial \vartheta} \Big|_{\vartheta=0} = 0$.

Taking the Laplace transform [137, p. 1020, 29.2.5] and rearranging the terms, one obtains

$$\frac{d^2\mathcal{U}(\xi; r)}{d\xi^2} - r(r + \sqrt{\tau_v})\mathcal{U}(\xi; r) = 0, \quad (\xi; r) \geq 0 \quad (4.1.3)$$

where $\mathcal{U}(\xi; r)$ is the Laplace transform of the excess velocity.

General solution for the equation (4.1.3) is

$$\mathcal{U}(\xi; r) = C_1(r)\exp\left\{-\xi[r(r + \sqrt{\tau_v})]^{\frac{1}{2}}\right\} + C_2(r)\exp\left\{\xi[r(r + \sqrt{\tau_v})]^{\frac{1}{2}}\right\}, \quad (4.1.4)$$

$(\xi; r) \geq 0$

Physically this solution is required to be bounded at $\xi \rightarrow \infty$, therefore, $C_2(r)$ must be identically zero. Denoting $C_1(r) \equiv C(r)$, we obtain

$$\mathcal{U}(\xi; r) = C(r)\exp\left\{-\xi[r(r + \sqrt{\tau_v})]^{\frac{1}{2}}\right\} \quad (4.1.5)$$

We are now able to eliminate the arbitrary parameter $C(r)$ by taking derivative of equation (4.1.5) with respect to ξ :

$$\frac{d\mathcal{U}}{d\xi} = -[r(r + \sqrt{\tau_v})]^{\frac{1}{2}}\mathcal{U} \quad (4.1.6)$$

which must be written as

$$-\mathfrak{U} = [r(r + \sqrt{\tau_v})]^{-\frac{1}{2}} \frac{d\mathfrak{U}}{d\xi} \quad (4.1.7)$$

The inverse Laplace transform of $[r(r + \sqrt{\tau_v})]^{-\frac{1}{2}}$ is simply [137, p. 1028, 29.3.124]

$$I_0\left(\frac{\vartheta}{2\sqrt{\tau_v}}\right) \exp\left(-\frac{\vartheta}{2\sqrt{\tau_v}}\right) \quad (4.1.8)$$

where $I_0(z)$ is the modified Bessel function of the first kind [137]. Taking the Laplace transform of (4.1.8) and applying the convolution theorem [20], we obtain

$$U_e = - \int_0^{\frac{t}{\sqrt{\tau}}} \frac{dU}{d\xi} I_0\left(\frac{\vartheta - \zeta}{2\sqrt{\tau_v}}\right) \exp\left(-\frac{\vartheta - \zeta}{2\sqrt{\tau_v}}\right) d\zeta, \quad t \geq 0 \quad (4.1.9)$$

After fully restoring the variables and rearranging the terms, equation (4.1.9) transforms into

$$u(y, t) = u_0 - \sqrt{\frac{v}{\tau_v}} \int_0^t \frac{\partial u(y, t^*)}{\partial y} I_0\left(\frac{t - t^*}{2\tau_v}\right) \exp\left(-\frac{t - t^*}{2\tau_v}\right) dt^*, (y, t) \geq 0 \quad (4.1.10)$$

which gives the relationship between the velocity and its spatial derivative at any moment of time and at any location in the domain in question.

The relation for shear stress is simply [16]

$$s(t + \tau_v) = -\mu \frac{\partial u(y, t)}{\partial y} \quad (4.1.11)$$

where μ is the dynamic viscosity of the fluid and s denotes shear stress. Also, $\mu = \rho \nu_k$, where ρ is the density of the fluid. Expanding equation (4.1.11) to Taylor series, we obtain

$$\frac{\partial u}{\partial y} = -\frac{1}{\rho \nu_k} \left(s + \tau_v \frac{\partial s}{\partial t} \right) \quad (4.1.12)$$

Hence, equation (4.1.10) for the fluid velocity takes the form

$$u(y, t) = u_0 - \sqrt{\frac{1}{\rho^2 \nu_k \tau_v}} \int_0^t \left[s(y, t^*) + \tau_v \frac{\partial s}{\partial t}(y, t^*) \right] I_0 \left(\frac{t - t^*}{2\tau_v} \right) \exp \left(-\frac{t - t^*}{2\tau_v} \right) dt^*, (y, t) \geq 0 \quad (4.1.13)$$

which can be rewritten in the following way

$$u(y, t) = u_0 - \sqrt{\frac{1}{\rho^2 \nu_k \tau_v}} \int_0^t \frac{\partial u(y, \zeta)}{\partial y} K_w \left(\frac{t - \zeta}{2\tau_v} \right) d\zeta \quad (4.1.14)$$

where the kernel K_w is

$$K_w(z) = I_0(z) \exp(-z) \quad (4.1.15)$$

However, if we want to apply a classical model here instead, then a different value of the kinematic viscosity ν_{eff} (let us call it effective viscosity) has to be assumed in order to match the experimental results. Thus,

$$u(y, t) = u_0 - \sqrt{\nu_{eff}(\tau_v)} \int_0^t \frac{\partial u(y, \zeta)}{\partial y} K(t - \zeta) d\zeta \quad (4.1.16)$$

where the kernel is given by

$$K(z) = \frac{1}{\sqrt{\pi z}} \quad (4.1.17)$$

Equating both equation (4.1.14) and equation (4.1.16), one obtains

$$\int_0^t \frac{\partial u}{\partial y} \left[\sqrt{\nu_{eff}} K(t - \zeta) - \sqrt{\frac{\nu_k}{\tau_v}} K_w \left(\frac{t - \zeta}{2\tau_v} \right) \right] d\zeta = 0 \quad (4.1.18)$$

or, as equation (4.1.18) should be valid for any $t > 0$,

$$\sqrt{\nu_{eff}} K(t) = \sqrt{\frac{\nu_k}{\tau_v}} K_w \left(\frac{t}{2\tau_v} \right) \quad (4.1.19)$$

Hence,

$$\nu_{eff} = \frac{\nu_k}{\tau_v} \left[\frac{K_w \left(\frac{t}{2\tau_v} \right)}{K(t)} \right]^2 \quad (4.1.20)$$

Restoring the original variables, one obtains that $K_w\left(\frac{t}{2\tau_v}\right) = I_0\left(\frac{t}{2\tau_v}\right) \exp\left(-\frac{t}{2\tau_v}\right)$ and

$K(t) = \frac{1}{\sqrt{\pi t}}$. Hence, we get

$$v_{eff} = \frac{v_k}{\tau_v} \left[\sqrt{\pi t} \cdot I_0\left(\frac{t}{2\tau_v}\right) \exp\left(-\frac{t}{2\tau_v}\right) \right]^2 \quad (4.1.21)$$

which is simply

$$v_{eff} = 2\pi v_k \left[\sqrt{\frac{t}{2\tau_v}} I_0\left(\frac{t}{2\tau_v}\right) \exp\left(-\frac{t}{2\tau_v}\right) \right]^2 \quad (4.1.22)$$

Let us now define $z = \frac{t}{2\tau_v}$, which, upon substituting into equation (4.1.22), gives us

the expression for the effective viscosity:

$$v_{eff} = 2\pi z v_k [I_0(z) \exp(-z)]^2 \quad (4.1.23)$$

The same mathematical procedure can be done for the one-dimensional thermal wave equation, which was done previously by Kulish and Novozhilov [12]:

$$\tau_\alpha \frac{\partial^2 T}{\partial t^2} + \frac{\partial T}{\partial t} = \alpha \frac{\partial^2 T}{\partial y^2} \quad (4.1.24)$$

where τ_α is the time lag due to thermal effects (relaxation time in phonon collisions)

and α stands for thermal diffusivity, both assumed to be constant. τ_α can be defined as

$$\tau_\alpha = \frac{3\alpha}{c^2} \quad (4.1.25)$$

where c is the speed of sound [12].

Thus, the relationship between the temperature and its spatial derivative at any moment of time and any location in the domain in question can be obtained [12]:

$$T(y, t) = T_0 - \left(\frac{\alpha}{\tau}\right)^{1/2} \int_0^t \frac{\partial T(y, t')}{\partial y} I_0 \left(\frac{t - t'}{2\tau_\alpha} \right) \exp \left(-\frac{t - t'}{2\tau_\alpha} \right) dt' \quad (4.1.26)$$

In the same manner, the expression for the apparent thermal diffusivity can be derived, which was previously done by Kulish and Chan [15]. They claimed that the increase of apparent thermal diffusivity could exceed by almost three orders of magnitude; however, it is not true due to a computational error in their solution.

The proper expression for the effective thermal diffusivity α_{eff} should be

$$\alpha_{eff} = 2\pi\alpha z [I_0(\bar{z}) \exp(-\bar{z})]^2 \quad (4.1.27)$$

where $\bar{z} = \frac{t}{2\tau_\alpha}$, which is different from the result obtained by Kulish and Chan. This formula shows that an apparent increase of thermal diffusivity really takes place but only exceeds the normal one by approximately 1.4 times only.

As we know, $\tau_\alpha = \frac{3\alpha}{c^2}$, $\tau_v = \frac{3\nu_k}{c^2}$, hence,

$$\frac{\tau_v}{\tau_\alpha} = \frac{\nu_k}{\alpha} = \text{Pr} \quad (4.1.28)$$

where Pr denotes the Prandtl number. Therefore, $\tau_v = \text{Pr} \cdot \tau_\alpha$ and $\bar{z} = \text{Pr} \cdot z$.

The numerical simulation results for the effective viscosity:

$$\frac{\nu_{eff}}{\nu_k} = 2\pi z [I_0(z) \exp(-z)]^2$$

Figure 4.1. Numerical simulation for the apparent viscosity behavior.

This graph shows the increase of the effective viscosity approximately to 1.4 times, which then drops again to its normal value.

Numerical results for the effective thermal diffusivity:

$$\frac{\alpha_{eff}}{\alpha} = 2\pi \bar{z} [I_0(\bar{z}) \exp(-\bar{z})]^2$$

Figure 4.2.

Numerical simulation for the apparent thermal diffusivity behavior.

This function shows that the value of effective thermal diffusivity increases in approximately 1.4 times and then returns to the standard value.

Let us now express the equation for thermal diffusivity in terms of variable z instead of $\bar{z}$. Thus, we obtain:

$$\begin{aligned} \frac{\nu_{eff}}{\nu_k} &= 2\pi z [I_0(z) \exp(-z)]^2 \\ \frac{\alpha_{eff}}{\alpha} &= 2\pi \text{Pr} z [I_0(\text{Pr} \cdot z) \exp(-\text{Pr} \cdot z)]^2 \end{aligned} \quad (4.1.29)$$

Now we are assuming that the fluid under consideration is a gas, for most of which Prandtl number is around 0.75. Numerical simulation for this case shows that

Figure 4.3.

Numerical simulation for apparent thermal diffusivity and viscosity for $Pr=0.75$.

- for a mixture of noble gases with hydrogen, $Pr=0.16$:

Figure 4.4.

Numerical simulation for apparent thermal diffusivity and viscosity for $Pr=0.16$.

- for mercury, $Pr=0.015$:

Figure 4.5.

Numerical simulation for apparent thermal diffusivity and viscosity for $Pr=0.015$.

- for liquid Helium I ($T=2.178K$), $Pr=0.6139$:

Figure 4.6.

Numerical simulation for apparent thermal diffusivity and viscosity for $Pr=0.6139$.

- for water, $Pr=7$:

Figure 4.7.

Numerical simulation for apparent thermal diffusivity and viscosity for $Pr = 7$.

- for $Pr=20$:

Figure 4.8.

Numerical simulation for apparent thermal diffusivity and viscosity for $Pr = 20$.

As we can see from the two previous pictures, when apparent thermal diffusivity reaches its peak, apparent viscosity is still on its increase stage and can be much smaller than its normal value. Thus, for bigger values of Prandtl number (when convective mode of heat transfer is dominant and which is common for engine oils and lubricants) the distance between two peaks of $\frac{\nu_{eff}}{\nu_k}$ and $\frac{\alpha_{eff}}{\alpha}$ also becomes bigger which can be considered as “superfluid-like” behaviour of fluid.

4.2 Steady State Heat Transfer within a Nanoscale Spatial Domain

According to numerous studies [138-141], the thermal conductivity in ultra-thin films is size-dependent, which means that the thermal conductivity in such a length scale becomes strictly a function of a spatial variable. In order to define that function, we consider the relationship between the local temperature and its spatial derivative within an one-dimensional domain, assuming that the finite speed of thermal waves propagation in isotropic and homogeneous medium, which takes the following form [12]

$$T(x, t) = T_0 - \sqrt{\frac{\alpha}{\tau}} \int_0^t \frac{\partial T(x, \zeta)}{\partial x} K_w \left(\frac{t - \zeta}{2\tau} \right) d\zeta, \quad (4.2.1)$$

where α is the thermal diffusivity, τ stands for the relaxation time, T_0 is the initial temperature and the kernel is $K_w(z) = I_0(z) \exp(-z)$ (as stated in equation (4.1.15)). Again, we wish to describe the behaviour of temperature in equation (4.2.1) by the classical model of heat transfer. For the classical model, the relationship between the local temperature and its spatial derivative within a one-dimensional semi-infinite domain [142] is

$$T(x, t) = T_0 - \sqrt{\alpha_{eff}} \int_0^t \frac{\partial T(x, \zeta)}{\partial x} K(t - \zeta) d\zeta, \quad (x, t) \geq 0 \quad (4.2.2)$$

where $K(z) = \frac{1}{\sqrt{\pi z}}$. These two equations combined would give us the relation for the effective thermal diffusivity as was stated in the previous section:

$$\alpha_{eff} = \alpha \frac{\pi t}{\tau} \left[I_0 \left(\frac{t}{2\tau} \right) \exp \left(-\frac{t}{2\tau} \right) \right]^2 \quad (4.2.3)$$

The effective thermal diffusivity given by equation (4.2.3) allows us to describe the solution of the hyperbolic heat equation by means of the classical parabolic heat equation.

We now are able to write the relationship between the time and spatial variables using the fact that the speed of thermal wave propagation is equal to the speed of sound in the transporting medium [143], as follows

$$t = \frac{x}{c} \quad (4.2.4)$$

where x is the spatial variable (which can never be too large due to the real domain being finite) and c is the speed of sound in the transporting medium. Substituting equation (4.2.4) into equation (4.2.3), the effective thermal diffusivity can be now written as a function of the spatial variable in the following way

$$\alpha_{eff} = \alpha \frac{\pi x}{\delta} \left[I_0 \left(\frac{x}{2\delta} \right) \exp \left(-\frac{x}{2\delta} \right) \right]^2 \quad (4.2.5)$$

where $\delta = \tau c$ is the characteristic length traveled by thermal wave. We now introduce the following dimensionless variables

$$\bar{\alpha} = \frac{\alpha_{eff}}{\alpha}, \eta = \frac{x}{\delta} \quad (4.2.6)$$

Now let us rewrite equation (4.2.5) in the dimensionless form:

$$\bar{\alpha} = \pi\eta \left[I_0 \left(\frac{\eta}{2} \right) \exp \left(-\frac{\eta}{2} \right) \right]^2 \quad (4.2.7)$$

The relationship between the thermal diffusivity and thermal conductivity is

$$k = c_p \rho \alpha \quad (4.2.8)$$

where c_p is the specific heat, ρ stands for the density of the medium, which both are assumed to be constant in our case. We now can write an equation for the size-dependent thermal conductivity in a dimensionless form combining equation (4.2.7) and equation (4.2.8) in the following form

$$\bar{k} = \pi\eta \left[I_0 \left(\frac{\eta}{2} \right) \exp \left(-\frac{\eta}{2} \right) \right]^2 \quad (4.2.9)$$

where $\bar{k} = \frac{k_{eff}}{k}$ denotes the dimensionless thermal conductivity, k_{eff} is the effective thermal conductivity of the medium. Hence, the equation (4.2.9) provides the phenomenological relationship between the dimensionless thermal conductivity and the dimensionless spatial variable.

Figure 4.9 represents that the dimensionless thermal conductivity from equation (4.2.9) decreases, when $\log(\eta)$ is less than zero, and, when $\log(\eta)$ tends to infinity, the dimensionless thermal conductivity $\bar{k} = \frac{k_{eff}}{k}$ becomes equal to one. In other words, the effective thermal conductivity becomes less than the bulk value in case, when the size of the transporting medium is less than the characteristic length of this medium. And when the size is much larger than the characteristic length, the effective thermal conductivity becomes the same as the bulk thermal conductivity. Hence, the point $\eta = 1$, at which the size of the transporting medium is equal to the characteristic length, becomes a critical point, beyond or below which the value of the thermal conductivity $\bar{k}$ is decreased.

Fig. 4.9.

Dependence of the thermal conductivity on the logarithmic-scale spatial variable in dimensionless form:

$$\eta_{max} = 1.53 \text{ and } \bar{k}_{max} = 1.38.$$

The function defined by the equation (4.2.6) has the maximum (see Fig. 4.9), to find which we consider

$$\frac{d\bar{k}}{d\eta} = \pi K_w(\eta)^2 + \pi\eta[K_1(\eta)K_w(\eta) - K_w(\eta)^2] = 0 \quad (4.2.10)$$

where $K_1(\eta) = I_1\left(\frac{\eta}{2}\right)$ and $I_1(z)$ is the modified Bessel function of the first kind of the first order [137, p. 355]. The solution of the equation (4.2.10) is

$$\eta_{max} = 1.53 \quad (4.2.11)$$

Upon substitution of equation (4.2.11) to equation (4.2.9) we can find the maximum value of the dimensionless thermal conductivity

$$\bar{k}_{max} = 1.38 \quad (4.2.12)$$

Hence, the size-dependent thermal conductivity in equation (4.2.9) has a maximum, when the size of the transporting medium becomes 1.53 times larger than the characteristic length of this medium, and the maximal value of the thermal conductivity is about 40 percent larger than its bulk value.

Let us now compare our theoretical results given by the equation (4.2.9) with the size-dependent thermal conductivity of silicon thin films obtained from experiments [144-146]. The parameters of silicon used in the equation (4.2.9) are shown in Table 4.1.

	$c(\text{s/m})$	$\tau(\text{ms})$	$\delta(\text{nm})$
Silicon ¹	6400	2.6	409

Table 4.1.

Room-temperature parameters of Si used in equation (4.2.9) for prediction [147].

The comparison results are shown in Fig. 4.10, where the thermal conductivity of silicon thin films is changed with the thickness of thin films lying in the range of 10-250 nm. The analytical prediction is in excellent agreement with the experimental results.

Fig. 4.10.

(Solid curve) The line of the size-dependent thermal conductivity of Si thin film at 300 K as predicted from equation (4.2.9). The symbols are experimental results: the triangles, squares, and rhombus cited in Ref. [144], [145], and [146], respectively.

Now we consider a one-dimensional steady state heat transfer problem through a medium, the size of which can be changed up to the nanoscale level as shown in Fig. 4.11.

Fig. 4.11.

The problem statement.

The two media are the heater and cooler with temperatures T_2 and T_1 ($T_2 > T_1$), which are respectively applied to the top and bottom boundaries of the transporting medium. We can apply Fourier's law in this case

$$q''(x) = -k_{eff}(x) \frac{dT}{dx} \quad (4.2.13)$$

We now rewrite equation (4.2.13) using equations (4.2.6) and (4.2.9)

$$q''(\eta) = -\pi\eta \left[I_0\left(\frac{\eta}{2}\right) \exp\left(-\frac{\eta}{2}\right) \right]^2 \frac{dT}{d\eta} \quad (4.2.14)$$

Let us now approximate the temperature gradient as

$$\frac{dT}{d\eta} \approx \frac{\Delta T}{\Delta\eta} = \frac{\Delta T}{\eta - \eta_0} \quad (4.2.15)$$

We now substitute equation (4.2.15) to equation (4.2.14) and set $\eta_0 = 0$ to get

$$q''(x) = -\pi \left[I_0 \left(\frac{\eta}{2} \right) \exp \left(-\frac{\eta}{2} \right) \right]^2 \Delta T \quad (4.2.16)$$

or

$$q''(\eta) = -H(\eta) \Delta T \quad (4.2.17)$$

where is $H(x) = k \frac{\pi}{\delta} \left[I_0 \left(\frac{x}{2\delta} \right) \exp \left(-\frac{x}{2\delta} \right) \right]^2$ is the heat transfer coefficient (in dimensional form).

We now rewrite the function $H(x)$ in the dimensionless form in order to analyse the behaviour of the heat flux:

$$\bar{H}(\eta) = \frac{\delta H(\eta)}{k} = \pi \left[I_0 \left(\frac{\eta}{2} \right) \exp \left(-\frac{\eta}{2} \right) \right]^2$$

Figure 4.12 represents the dependence of the modified heat transfer coefficient $\bar{H}(\eta)$ on the logarithmic-scale spatial variable in a dimensionless form.

Fig. 4.12.

Dependence of the heat transfer coefficient on the logarithmic-scale spatial variable in dimensionless form.

The figure shows that there are two limits of the heat transfer coefficient: the upper limit equals π , and the lower limit is equal to zero. When the value of η tends to one, the value of the heat transfer coefficient is increased and then reaches the upper limit, when the η tends to zero.

Thus, the heat transport properties of the medium characterised by the modified heat transfer coefficient $\bar{H}$ are changed with size of the domain and when the size becomes comparable with the characteristic length of the medium under consideration, the heat transfer coefficient starts to increase and then, when the size continues to tend to zero, the heat transfer coefficient achieves its upper limit which is defined as

$$H_{upper} = \frac{\pi k}{\delta},$$

instead of infinity predicted by the classical Fourier law.

The study shows that the thermal conductivity decreases when the size of the transporting medium becomes less than the characteristic length. On the other hand, the value of the thermal conductivity is equal to its bulk value in the transporting medium when the size of this medium becomes at least one order of magnitude larger than the characteristic length. Combining the defined size- dependent thermal conductivity with Fourier's law allows us to study the behaviour of the heat flux within a nanoscale gap.

Chapter 5. Apparent Source

Apparent source/sink terms appear in heat transport equation, which has a source/sink term $S(t)$, after introducing the time lag τ . Hence, the resulting equation would contain the term $\tau \frac{\partial S(t)}{\partial t}$.

In order to study the apparent source/sink term, let us consider a one-dimensional time-dependent diffusion equation in a semi-infinite medium, with a space and time-dependent volumetric heat source or sink. The diffusion equation, assuming constant and uniform properties, is

$$\frac{\partial T(x, t)}{\partial t} - \alpha \frac{\partial^2 T(x, t)}{\partial x^2} - \frac{\dot{q}'''(x, t)}{\rho c_p} = 0, x \geq 0, t \geq 0 \quad (5.1.1)$$

where T stands for the scalar temperature, t is the time, $\dot{q}'''(x, t)$ denotes volumetric source or sink term (heat-power per unit-volume) and $\alpha = \frac{k_c}{\rho c_p}$ is the thermal diffusivity of the fluid. Equation (5.1.1) is now applied to solve the initial value problem in a semi-infinite domain under the condition of initial equilibrium of the medium in the domain ($T = T_0$ everywhere at $(x, t) = (0, 0)$, $T(0, t) = T_0$ and $\frac{\partial T}{\partial t}\bigg|_{t=0} = 0, \frac{\partial T}{\partial x}\bigg|_{x=0} = 0$). For the semi-infinite domain, when $x \rightarrow \infty$ the boundary condition is $T \rightarrow T_0$.

Introducing the change of variables $\xi = x\sqrt{\alpha}$ and $T^* = T - T_0$, equation (5.1.1) transforms into

$$\frac{\partial T^*(\xi, t)}{\partial t} - \frac{\partial^2 T^*(\xi, t)}{\partial \xi^2} - \frac{\dot{q}'''(\xi, t)}{\rho c_p} = 0 \quad (5.1.2)$$

The initial condition transforms into $T^*(\xi, 0) = 0$. Taking the Laplace transform [137, p.1020, 29.2.5] of equation (5.1.2) and using the initial condition, we obtain

$$\frac{d^2 \Theta}{d\xi^2} - s\Theta + Q(\xi, s) = 0 \quad (5.1.3)$$

where $\Theta(\xi, s)$ is the Laplace transform of $T^*(\xi, t)$, and $Q(\xi, s)$ is the Laplace transform of the $\frac{\dot{q}'''(\xi, t)}{\rho c_p}$. Equation (5.1.3) is the ordinary differential equation, which is known as the forced oscillations equation. The solution to this nonhomogeneous equation can be written as

$$\Theta(\xi, s) = C_1(s)\exp\left[\xi s^{1/2}\right] + C_2(s)\exp\left[-\xi s^{1/2}\right] + P(\xi, s) \quad (5.1.4)$$

The first two terms on the right side form the general solution to the associated homogeneous equation (when $Q(\xi, s) = 0$) and $P(\xi, s)$ is the particular solution.

The solution of equation (5.1.4) has to be bounded as $\xi \rightarrow \infty$, hence, the arbitrary parameter $C_1(s)$ has to be zero; so, the equation (5.1.4) becomes

$$\Theta(\xi, s) = C(s)\exp\left[-\xi s^{1/2}\right] + P(\xi, s) \quad (5.1.5)$$

where $C(s) = C_2(s)$.

The constant $C(s)$ can be rewritten as

$$C(s) = [\Theta(\xi, s) - P(\xi, s)]\exp\left[\xi s^{1/2}\right] \quad (5.1.6)$$

Let us now differentiate equation (5.1.5) with respect to ξ :

$$\frac{\partial\Theta(\xi, s)}{\partial\xi} = -C(s)s^{1/2}\exp\left[\xi s^{1/2}\right] + \frac{\partial P(\xi, s)}{\partial\xi} \quad (5.1.7)$$

Now let us eliminate constant $C(s)$ combining equations (5.1.6) and (5.1.7):

$$\frac{\partial\Theta(\xi, s)}{\partial\xi} = -s^{1/2}\Theta(\xi, s) + s^{1/2}P(\xi, s) + \frac{\partial P(\xi, s)}{\partial\xi} \quad (5.1.8)$$

Applying the inverse Laplace transform on equation (5.1.7) and restoring the original variables, we obtain [137, p. 1020, 29.2.5; p.1025, 29.3.63]:

$$\begin{aligned} \frac{\partial T(x, t)}{\partial x} = \frac{1}{\sqrt{\alpha}} & \left[\frac{T_0}{\sqrt{\pi t}} - \frac{\partial^{1/2} T(x, t)}{\partial t^{1/2}} + \frac{\partial^{1/2} p(x, t)}{\partial t^{1/2}} - \frac{p(x, 0)}{\sqrt{\pi t}} \right] \\ & + \frac{\partial p(x, t)}{\partial x} \end{aligned} \quad (5.1.9)$$

where $p(x, t)$ is the inverse Laplace transform of the function $P(x, s)$ and partial

derivative operator is defined as $\frac{\partial^\gamma}{\partial x^\gamma} g(x) = \frac{1}{\Gamma(\gamma)} \int_0^x (x-t)^{\gamma-1} g(t) dt$, which is

related to the Abel's integral equations in the following way: $w(x) = \Gamma(1 - \gamma)g^{\gamma-1}(j(x))$, where $w(x)$ is a continuous function and both $w(x)$ and $j(x)$ satisfy Abel's integral equations of first and second kind [148]: $w(x) = \int_0^x \frac{j(t)}{\sqrt{x-t}} dt$ and $j(x) = w(x) + \int_0^x \frac{j(t)}{\sqrt{x-t}} dt$.

Invoking a Fourier law with the equation (5.1.9), we get an expression, which relates the heat flux $\dot{q}''(x, t)$ and the temperature $T(x, t)$ in any location inside domain:

$$\dot{q}''(x, t) = \frac{k_c}{\sqrt{\alpha}} \left[-\frac{T_0}{\sqrt{\pi t}} + \frac{\partial^{1/2} T(x, t)}{\partial t^{1/2}} - \frac{\partial^{1/2} p(x, t)}{\partial t^{1/2}} + \frac{p(x, 0)}{\sqrt{\pi t}} \right] - k_c \frac{\partial p(x, t)}{\partial x} \quad (5.1.10)$$

Now, let us derive the expression for the local temperature $T(x, t)$ using the fractional operator $\frac{\partial^{-1/2}(\cdot)}{\partial t^{-1/2}}$ and the following properties of the fractional calculus [20]

$$\begin{aligned} \frac{\partial^h}{\partial t^h} \left(\frac{\partial^f g(t)}{\partial t^f} \right) &= \frac{\partial^{f+h} g(t)}{\partial t^{f+h}}; \quad \frac{\partial^f [t^n]}{\partial t^f} = \frac{\Gamma(n+1)t^{n-f}}{\Gamma(n+1-f)}; \\ \frac{\partial^{-1/2}(C)}{\partial t^{-1/2}} &= 2C \sqrt{\frac{t}{\pi}} \end{aligned} \quad (5.1.11)$$

The result is

$$\begin{aligned} T(x, t) &= T_0 - p(x, 0) + p(x, t) \\ &+ \frac{\sqrt{\alpha}}{k_c} \frac{\partial^{-1/2} \left[\dot{q}''(x, t) + k_c \frac{\partial p(x, t)}{\partial x} \right]}{\partial t^{-1/2}} \end{aligned} \quad (5.1.12)$$

Equation (5.1.12) contains terms depending on two different groups of thermal properties, namely, $\sqrt{\alpha}$ and k_c . Hence, equation (5.1.12) can be rewritten as

$$T(x, t) = T_0 + T_f(x, t) + T_v(x, t) \quad (5.1.13)$$

where the last two terms are the contributions from the local heat flux and the local volumetric heat source or sink term, respectively, which have the following form:

$$T_f(x, t) = \frac{\sqrt{\alpha}}{k_c} \frac{\partial^{-1/2} [\dot{q}''(x, t)]}{\partial t^{-1/2}} \quad (5.1.14)$$

And

$$T_v(x, t) = -p(x, 0) + p(x, t) + \sqrt{\alpha} \frac{\partial^{-1/2} \left[\frac{\partial p(x, t)}{\partial x} \right]}{\partial t^{-1/2}} \quad (5.1.15)$$

Now, let us consider the case of the apparent source/sink term $\tau \frac{\partial \dot{q}'''(x, t)}{\partial t}$ added to equation (5.1.1), where τ is the phase lag time.

As we know the general solution to equation (5.1.1) without source/sink term, to obtain a solution containing this term we need to find its particular solution and add it to the general one. If the heating area is much bigger than the probing area and the properties of the material are considered uniform and constant, the energy balance equation can be considered unidirectional. Let us assume the radiation

power flux distribution at the surface has the form of Gaussian pulse (normal distribution):

$$I''(0, t) = I_b'' \exp \left[- \left(\frac{t - b}{\sigma} \right)^2 \right] \quad (5.1.16)$$

where I_b'' is the incidence radiation power flux at $t = b$ and equals $\frac{F}{\sigma\sqrt{\pi}}$, with F being the fluence of the laser irradiation (i.e. radiation energy E per unit of heated surface A), b and $\frac{\sigma}{\sqrt{2}}$ represent the mean value and the standard deviation of the normal distribution, respectively.

Then, let us assume that the material is semi-transparent. Hence, the boundary heat flux is set as equal to zero and the laser energy is assumed to be absorbed volumetrically within the material. The energy balance is identical to (5.1.1) with $\dot{q}'''(x, t)$ and its apparent term $\tau \frac{\partial \dot{q}'''(x, t)}{\partial t}$, the absorbed laser irradiation per unit volume, expressed as the Beer's law [149]

$$\dot{q}'''(x, t) = I_b''(1 - R) \exp(-\kappa x) \exp \left[- \left(\frac{t - b}{\sigma} \right)^2 \right] \quad (5.1.17)$$

where κ is the extinction or attenuation coefficient of the material assumed to be constant and R is the surface reflectivity. The light is assumed to be monochromatic, and no chemical reaction and radiation scattering takes place during the process. The fluid is homogeneous in the interaction volume and the incident flux does not influence atoms or molecules. The apparent term for the given laser pulse is

$$\tau \frac{\partial \dot{q}'''(0, t)}{\partial t} = -2\tau \left(\frac{t-b}{\sigma^2} \right) I_b'' \exp \left[- \left(\frac{t-b}{\sigma} \right)^2 \right] \quad (5.1.18)$$

giving the absorbed laser irradiation as

$$\tau \frac{\partial \dot{q}'''(x, t)}{\partial t} = -2\tau \left(\frac{t-b}{\sigma^2} \right) I_b'' (1-R) \exp(-\kappa x) \exp \left[- \left(\frac{t-b}{\sigma} \right)^2 \right] \quad (5.1.19)$$

and

$$\begin{aligned} \dot{q}'''(x, t) + \tau \frac{\partial \dot{q}'''(x, t)}{\partial t} = \\ I_b'' (1-R) \exp(-\kappa x) \left[1 - 2\tau \left(\frac{t-b}{\sigma^2} \right) \right] \exp \left[- \left(\frac{t-b}{\sigma} \right)^2 \right] \end{aligned} \quad (5.1.20)$$

Let us divide the equation (5.1.20) into two parts, namely, $\dot{q}_1'''(x, t)$ and $\dot{q}_2'''(x, t)$:

$$\dot{q}_1'''(x, t) = \left[1 + \frac{2\tau b}{\sigma^2} \right] I_b'' (1-R) \exp(-\kappa x) \exp \left[- \left(\frac{t-b}{\sigma} \right)^2 \right] \quad (5.1.21)$$

$$\dot{q}_2'''(x, t) = -\frac{2\tau t}{\sigma^2} I_b'' (1-R) \exp(-\kappa x) \exp \left[- \left(\frac{t-b}{\sigma} \right)^2 \right], (x, t) > 0 \quad (5.1.22)$$

The Laplace transform [137, p. 1028, 29.3.112] of equation (5.1.21), after dividing it by ρc_p , and using $\xi = \frac{x}{\sqrt{\alpha}}$, is

$$Q_1(\xi; s) = \left[1 + \frac{2\tau b}{\sigma^2}\right] Q_p(\xi; s) \quad (5.1.23)$$

where

$$Q_p(\xi; s) = \frac{I_b''}{2\rho c_p} (1 - R) \kappa \sigma \sqrt{\pi} \exp[-\kappa \sqrt{\alpha} \xi] \exp\left[\frac{\sigma^2}{4s^2 - bs}\right] \times \operatorname{erfc}\left[\frac{\sigma}{2}\left(s - \frac{2b}{\sigma^2}\right)\right], \quad (5.1.24)$$

where $\operatorname{erfc}(x) = 1 - \operatorname{erf}(x) = \frac{2}{\sqrt{\pi}} \int_x^\infty e^{-t^2} dt$ is the complementary error function

and $\operatorname{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-t^2} dt$ is the error function.

Taking the same Laplace transform [137, p. 1028, 29.3.112] of equation (5.1.22),

we obtain:

$$Q_2(\xi; s) = \frac{2\tau b}{\sigma^2} \frac{\partial Q_p(\xi; s)}{\partial s} \quad (5.1.25)$$

Then, the full Laplace transform of $\dot{q}'''(x, t) + \tau \frac{\partial \dot{q}'''(x, t)}{\partial t}$ is

$$Q(\xi; s) = Q_1(\xi; s) + Q_2(\xi; s) \quad (5.1.26)$$

The particular solution is

$$P(\xi; s) = \frac{\tilde{Q}(s)}{s - \kappa^2 \alpha} \exp[-\kappa \sqrt{\alpha} \xi] \quad (5.1.27)$$

where $\tilde{Q}(s) = Q(\xi; s) \exp[\kappa \sqrt{\alpha} \xi] = \{Q_1(\xi; s) + Q_2(\xi; s)\} \exp[\kappa \sqrt{\alpha} \xi]$. To get the particular solution of the original problem, we need to find the inverse Laplace transform of the expression (5.1.27), which can be found in two steps. Let us write (5.1.27) in the following way:

$$\begin{aligned} P(\xi; s) &= P_1(\xi; s) + P_2(\xi; s); \\ P_1(\xi; s) &= \frac{Q_1(\xi; s)}{s - \kappa^2 \alpha} \exp[-\kappa \sqrt{\alpha} \xi]; \\ P_2(\xi; s) &= \frac{Q_2(\xi; s)}{s - \kappa^2 \alpha} \exp[-\kappa \sqrt{\alpha} \xi]. \end{aligned} \quad (5.1.28)$$

Taking into account (5.1.23), we get

$$P_1(\xi; s) = \left[1 + \frac{2\tau b}{\sigma^2} \right] \frac{Q_p(\xi; s)}{s - \kappa^2 \alpha} \exp[-\kappa \sqrt{\alpha} \xi] \quad (5.1.29)$$

By applying convolution theorem and taking into account that the inverse Laplace transform of $Q_p(\xi; s) \exp[\kappa \sqrt{\alpha} \xi]$ is $\frac{I_b''}{2\rho c_p} (1 - R) \kappa \sigma \sqrt{\pi} \kappa \exp \left[- \left(\frac{t-b}{\sigma} \right)^2 \right]$ [137, p. 1028, 29.3.112], and the inverse Laplace transform of $\frac{1}{s - \kappa^2 \alpha}$ is $\exp[-\kappa \sqrt{\alpha} t]$, we obtain for $P_1(\xi; s)$

$$p_1(x, t) = \left[1 + \frac{2\tau b}{\sigma^2}\right] \hat{p}(x, t) \quad (5.1.30)$$

where

$$\begin{aligned} \hat{p}(x, t) = & \frac{I_b''}{2\rho c_p} (1 - R) \kappa \sigma \sqrt{\pi} \exp[-\kappa x] \times \exp[\kappa^2 \alpha t] \times \exp\left[\frac{-b^2}{\sigma^2 + K^2}\right] \\ & \times \left[\operatorname{erf}\left(\frac{t}{\sigma} + K\right) - \operatorname{erf}(K)\right] \end{aligned} \quad (5.1.31)$$

where

$$K = \frac{\kappa^2 \alpha \sigma^2 - 2b}{2\sigma} \quad (5.1.32)$$

For the second term $P_2(\xi; s)$, the inverse Laplace transform [137, p. 1028, 29.3.112] equals

$$p_1(x, t) = -\frac{2\tau t}{\sigma^2} \hat{p}(x, t) \quad (5.1.33)$$

Hence, the full inverse Laplace transform of the particular solution $P(\xi; s)$ is

$$p(x, t) = p_1(x, t) + p_2(x, t) = \left[1 + \frac{2\tau}{\sigma^2}(t - b)\right] \hat{p}(x, t) \quad (5.1.34)$$

Noticing that $\frac{\partial p(x, t)}{\partial x} = -\kappa p(x, t)$, we obtain:

$$p(0, t) = \left[1 + \frac{2\tau}{\sigma^2}(t - b)\right] \frac{I_b'' \alpha \sigma \kappa}{2\sqrt{\pi}} \left[\operatorname{erf}\left(\frac{b}{\sigma}\right) \exp[\kappa^2 \alpha t] - \operatorname{erf}\left(\frac{b-t}{\sigma}\right) \right] \quad (5.1.35)$$

and,

$$\begin{aligned} \frac{\partial p(0, t)}{\partial x} = & \left[1 + \frac{2\tau}{\sigma^2}(t - b)\right] \frac{I_b'' \alpha \sigma \kappa^2}{2\sqrt{\pi}} \left[-\operatorname{erf}\left(\frac{b}{\sigma}\right) \exp[\kappa^2 \alpha t] \right. \\ & \left. + \operatorname{erf}\left(\frac{b-t}{\sigma}\right) \right] \end{aligned} \quad (5.1.36)$$

In order to get the contribution from volumetric source term and apparent source term, we put $p(0, 0) = 0$ and obtain the following expression:

$$T_v(0, t) = A^* \left[D^* - \frac{\kappa}{\sqrt{\alpha}} \frac{\partial^{-1/2} D^*}{\partial t^{-1/2}} \right] \quad (5.1.37)$$

where $A^* = \frac{I_b'' \alpha \sigma \kappa}{2\sqrt{\pi}}$ and $D^* = \left[1 + \frac{2\tau}{\sigma^2}(t - b)\right] \left[\operatorname{erf}\left(\frac{b}{\sigma}\right) \exp[\kappa^2 \alpha t] - \operatorname{erf}\left(\frac{b-t}{\sigma}\right) \right]$.

Now we wish to impose this solution to the flow motion equation, which has a similar form

$$\frac{\partial u(x, t)}{\partial t} = \nu_k \frac{\partial^2 u(x, t)}{\partial x^2} + \frac{S(t)}{\rho} \quad (5.1.38)$$

with similar boundary and initial conditions. We are solving the initial value problem in a semi-infinite domain under the condition of initial equilibrium of the fluid in the domain ($u = u_0$ everywhere at time $t = 0$ and $\frac{\partial u}{\partial t}\bigg|_{t=0} = 0, \frac{\partial u}{\partial x}\bigg|_{x=0} = 0$). For the semi-infinite domain, when $x \rightarrow \infty$ the boundary condition is $u \rightarrow u_0$. Here ν_k is the kinematic viscosity of the fluid and $S(t)$ is volumetric source/sink term.

The apparent source/sink term here has the following form: $\tau \frac{\partial S(t)}{\partial t}$. We can use the similar mode of solving this equation in order to find a contribution to the velocity of the flow from the volumetric and apparent terms by doing respective changes in the constants. Hence, for the velocity component due to apparent source presence we obtain:

$$u_v(0, t) = A^* \left[D^* - \frac{\kappa}{\sqrt{\nu_k}} \frac{\partial^{-1/2} D^*}{\partial t^{-1/2}} \right] \quad (5.1.39)$$

where $A^* = \frac{I_b'' \sigma \nu_k \kappa}{2\sqrt{\pi}}$ and $D^* = \left[1 + \frac{2\tau}{\sigma^2} (t - b) \right] \left[\text{erf}\left(\frac{b}{\sigma}\right) \exp[\kappa^2 \nu_k t] - \text{erf}\left(\frac{b-t}{\sigma}\right) \right]$.

Chapter 6. Model Validation

6.1. Introduction

Understanding heat transfer processes in metallic films induced by ultrashort laser pulse is of great importance due to their wide applications in microelectronics [150], micro- and nano-electro mechanical devices, data storage devices [151, 152] and other devices. The two-temperature model (TTM) proposed by Anisimov et al. [153] is widely employed [154] in order to study (theoretically and numerically) the laser interaction with metallic films. General form of the TTM is two coupled nonlinear parabolic partial differential equations describing the interaction of two subsystems: electron and phonon (lattice) gasses. This model is based on the assumption that electrons and phonons stay in thermal equilibrium. In particular, that was in detail investigated by Tzou [59], who then proposed a macroscopic dual-phase-lag theory. This theory is a result of modification of the TTM to single partial differential equation describing the effect of microscopic phonon–electron interaction and diffusion.

Numerous experimental studies have been performed [61,155-161] in order to investigate the heating of metallic films by laser with picosecond and subpicosecond pulses. It was shown that a finite time is needed to reach the electron–phonon thermal equilibrium. It means that ultrashort laser pulse irradiation can transiently bring the metallic film to a strong electron–phonon nonequilibrium stage, in which the temperature of electron gas rises whereas the state of phonon (lattice) gas does not change. The strong electron–phonon

nonequilibrium stage in metallic films is a totally different thermal response of the metallic film compared to a prediction from the original TTM but can still be included within the framework of TTM. A hyperbolic two-step model was rigorously derived by Qiu and Tien [60] from the Boltzmann transport equation for electrons to reflect this nonequilibrium stage in films. Klossika et al. [162] extended the TTM by Cattaneo's law for the heat flux in the electron gas to account for a finite propagation speed of heat. In addition, Cattaneo's law was applied by Chen and Beraun [163] for both electron and phonon subsystems. Also the numerical analysis of the proposed model was performed, which, in general, invokes a set of four partial differential equations. The proposed models, as a rule, do not have analytical solutions and are numerically analysed. This is due to the fact of physical complexity of the ultrafast heat transfer mechanism in metallic films. To clarify the physical essence of this phenomenon, further theoretical and experimental investigations of developed models are required.

In this thesis, the results of experimental [164] and theoretical studies of heat transfer processes in thin gold films induced by 100 fs laser pulses with a time resolution of 50 fs are presented. The analysis of experimental results shows that the heat transfer in thin gold films occurs via electron-phonon and phonon-phonon interactions. The resulting temperature wave appears to be dissipated in a surface layer of thin film within 800 fs. Observed decay times of 1.6 to 0.8 ps correspond to the electron-phonon relaxation, and 12 ps corresponds to the phonon-phonon relaxation. Moreover, the electron-phonon relaxation time varies versus the probe wavelength; at the same time, the phonon-phonon relaxation time shows a weak dependence on the probe wavelength.

As the electron gas possesses small heat capacity, thermalization caused by electron–electron interactions [165, 166] takes place during a time scale less than a few hundred femtoseconds. This time is relatively small comparing with the total duration of the surface temperature response, which occurs in more than 10 ps. Thus, the heating of the electron gas is assumed to be instantaneous. After applying Cattaneo’s law to the phonon subsystem of TTM, it becomes possible to transform the model into the hyperbolic heat equation [167]. The mathematical model of this chapter takes into account both sources, i.e., the volumetric and surface heat flux. The process is studied analytically within a semi-infinite domain and agrees well with the experimental data. Using this model, we are able to define the time of electron–phonon relaxation as the ratio of the penetration depth to the speed of sound in the bulk material. The definition of the electron–phonon relaxation time has a physical meaning as the dependence of the penetration depth on the wavelength has been experimentally observed, for instance, in [168]. This definition also provides an explanation of experimental results, which show dependence of the electron–phonon relaxation time on the wavelength.

6.2 Mathematical Model

Generally, the TTM can be expressed in terms of two coupled nonlinear partial differential equations for one-dimensional problem as follows:

$$C_e(T_e) \frac{\partial T_e}{\partial t} = \frac{\partial}{\partial x} \left[k_e(T_e, T_{ph}) \frac{\partial T_e}{\partial x} \right] - G(T_e)(T_e - T_{ph}) + S(x, t) \quad (6.2.1)$$

$$C_{ph}(T_{ph}) \frac{\partial T_{ph}}{\partial t} = \frac{\partial}{\partial x} \left[k_{ph}(T_{ph}) \frac{\partial T_{ph}}{\partial x} \right] - G(T_e)(T_e - T_{ph}) \quad (6.2.2)$$

where C and k stay for the heat capacities and thermal conductivities of the electrons and phonons as denoted by subscripts e and ph , respectively; $G(T_e)$ denote the electron–phonon coupling factor, and $S(x, t)$ is defined as a source term describing the local energy deposition by the laser pulse. The model is based on the assumption that electrons and phonons are in thermal equilibrium. Investigations of the experimental data show that the thermalization of the electron gas due to electron–electron interactions occurs during a time scale less than a few hundred femtoseconds [165, 166]. This time span is relatively small comparing to the total duration of the surface temperature response, which takes more than 10 ps. Thus, the heating of the electron gas can be assumed to be instantaneous, which means that in equation (6.2.1) the terms $C_e(T_e) \frac{\partial T_e}{\partial t}$ and $\frac{\partial}{\partial x} \left[k_e(T_e, T_{ph}) \frac{\partial T_e}{\partial x} \right]$ do not have any significant effect on the process of the electron gas heating. Hence, (6.2.1) can be simplified to become

$$G(T_e)(T_e - T_{ph}) \approx S(x, t), x \geq 0, t \geq 0. \quad (6.2.3)$$

Equation (6.2.3) shows that equilibrium between electron and phonon gases occurs after the laser excitation.

Although material parameters C_{ph} , k_{ph} , and G normally are functions of temperature, in our study we treat them as constants due to the short time of the processes. As the relaxation time of the phonon gas in metal is about 10 ps, we extend equation (6.2.2) by applying Cattaneo's law for the heat flux in the phonon gas, which is

$$\tau \frac{\partial q''_{ph}}{\partial t} + q''_{ph} = -k_{ph} \frac{\partial T_{ph}}{\partial x} \quad (6.2.4)$$

where τ is the relaxation time, and q''_{ph} denotes the heat flux of the phonon gas corresponding to the gradient of T_p . Combining equation (6.2.4) together with equation (6.2.2) and accounting for the law of energy conservation, we obtain

$$\begin{aligned} & \tau \frac{\partial^2 T_{ph}}{\partial t^2} + \frac{\partial T_{ph}}{\partial t} = \\ & \alpha_p \frac{\partial^2 T_{ph}}{\partial x^2} + \frac{1}{C_p} \left[G(T_e)(T_e - T_{ph}) + \tau \frac{\partial}{\partial t} G(T_e - T_{ph}) \right], x \geq 0, t \geq 0 \end{aligned} \quad (6.2.5)$$

where $\alpha_{ph} = \frac{k_{ph}}{C_{ph}}$ stands for the thermal diffusivity of the phonon gas.

Substituting equation (6.2.3) into equation (6.2.5), we obtain a one-dimensional

hyperbolic heat equation [19] with the time-dependent volumetric heat source, which can be written as follows:

$$\tau \frac{\partial^2 T}{\partial t^2} + \frac{\partial T}{\partial t} = \alpha \frac{\partial^2 T}{\partial x^2} + \left[q_v + \tau \frac{\partial q_v}{\partial t} \right] \quad (6.2.6)$$

where $q_v = S(x, t)/C$ ($C \equiv C_{ph}$). Electron-phonon subsystems are at equilibrium due to the assumption that the electron gas is heated instantaneously. Hence, all parameters of equation (6.2.6) can be defined as for the bulk material, and the subscript ph can be removed from the equation.

Klossika studied the propagation of heat in metal by means of waves with a finite speed. However, he neglected the existence of the volumetric heat source in his work [162]. In the present case under consideration, the penetration depth is of the order of a few nanometres. Nevertheless, the basic term which defines the surface temperature response under our experimental conditions (subpicosecond irradiation) is not the surface heat flux, but the volumetric source. Moreover, the penetration depth, which is included in the volumetric source, is an important characteristic that is responsible for the relaxation time of electron–phonon scattering in thin metal films. Hence, the volumetric source should not be neglected in a realistic model.

Now, let us rewrite (6.2.6) in a dimensionless form for the further analysis:

$$\frac{\partial^2 \theta(\eta, \xi)}{\partial \xi^2} + 2 \frac{\partial \theta(\eta, \xi)}{\partial \xi} = \frac{\partial^2 \theta(\eta, \xi)}{\partial \eta^2} + \left[\tilde{q}(\eta, \xi) + \frac{1}{2} \frac{\partial \tilde{q}(\eta, \xi)}{\partial \xi} \right] \quad (6.2.7)$$

where $\xi = t/(2\tau)$, $\eta = x/(2\sqrt{\alpha\tau})$, and $\theta(\eta, \xi)$ and $\tilde{q}(\eta, \xi)$ are the dimensionless temperature and volumetric heat sources, respectively.

We assume that the light intensity $I(t)$ of the laser pulse is Gaussian and that the metallic film absorbs laser irradiation energy exponentially. After correcting the reflection at the surface, the simulation of the volumetric heating within the material looks as the volumetric source:

$$S(x, t) = \frac{1 - R'}{\kappa} \exp\left(-\frac{x}{\kappa}\right) I(t) \quad (6.2.8)$$

where R' is the reflectivity of the irradiated surface, and κ stands for the radiation penetration depth. The dimensionless temperature is defined here as

$$\theta(\eta, \xi) = \frac{C\rho\sqrt{\alpha\tau\pi}}{F(1 - R')} [T - T_0] \quad (6.2.9)$$

where F is the laser irradiation fluence and T_0 denotes the initial temperature.

Hence, the expression for the dimensionless volumetric source becomes

$$\tilde{q}(\eta, \xi) = \frac{1}{\xi_p \Delta\eta} \exp\left(-\frac{\eta}{\Delta\eta} - a \left[\frac{\xi - \xi_b}{\xi_p}\right]^2\right) \quad (6.2.10)$$

where $\xi_p = \frac{t_p}{2\tau}$, $\xi_b = \frac{t_b}{2\tau}$, $\Delta\eta = \kappa/\sqrt{\alpha\tau}$, a is a positive constant (e.g., for a Gaussian distribution, $a = 2.77$), and t_p is the FWHM of the laser pulse.

Now, we are going to obtain the solution to equation (6.2.7) using the method proposed in [169], which was further developed for finite domains [170] and for nonhomogeneous problems [142]. This method leads to a Volterra-type integral equation that relates the local values of temperature and the corresponding heat flux. The equation is valid in the entire domain and its boundaries.

The solution procedure follows the one of [142]; so, now let us take the Laplace transform of equation (6.2.7). We take into account that initial condition of the system described by equation (6.2.7) is the thermal equilibrium, hence,

$$\left. \frac{\partial \theta}{\partial \xi} \right|_{\xi=0} = 0 \text{ and } \theta|_{\xi=0} = 0, \text{ which leads to}$$

$$\frac{d^2 \Theta}{d\eta^2} - (2s + s^2)\Theta + \left[1 + \frac{1}{2}s\right] \tilde{Q} = 0 \quad (6.2.11)$$

where $\Theta(\eta, s)$ and $\tilde{Q}(\eta, s)$ are the Laplace transforms of $\theta(\eta, \xi)$ and $\tilde{q}(\eta, \xi)$, respectively. The general solution of the nonhomogeneous equation (6.2.11) is

$$Q(\eta, s) = C_1(s)e^{\lambda(s)\eta} + C_2(s)e^{-\lambda(s)\eta} + P(\eta, s) \quad (6.2.12)$$

where $\lambda(s) = \sqrt{2s + s^2}$. The two first terms on the right side of equation (6.2.11) are the general solution of the associated homogeneous equation with $\tilde{Q}(\eta, s) = 0$, and $P(\eta, s)$ is a particular solution of equation (6.2.12).

Note that the surface heat flux is induced by the electron gas after absorbing the laser energy. Hence, the boundary conditions become

$$\left. \frac{\partial \theta}{\partial \eta} \right|_{\eta=0} = -\tilde{q}'' - \frac{1}{2} \frac{\partial \tilde{q}''}{\partial \xi}, \quad \lim_{n \rightarrow \infty} \theta = 0 \quad (6.2.13)$$

where $\tilde{q}'' = \frac{1}{\xi_p} \exp\left(-a \left[\frac{\xi - \xi_b}{\xi_p}\right]^2\right)$ is the dimensionless form of the surface heat flux. Hence, the parameter $C_1(s)$ must be zero and equation (6.2.12) can be simplified into the following expression:

$$Q(\eta, s) = C(s)e^{\lambda(s)\eta} + P(\eta, s) \quad (6.2.14)$$

We need to eliminate $C(s)$ from equation (6.2.12). For this purpose, the derivative of $\theta(\eta, s)$ with respect to η is used:

$$\frac{dQ(\eta, s)}{d\eta} = -\lambda(s)C(s)e^{-\lambda(s)\eta} + \frac{dP(\eta, s)}{d\eta} \quad (6.2.15)$$

Then, combining equation (6.2.14) and equation (6.2.15), and using the relationship between the temperature gradient and the surface heat flux equation (6.2.13), we obtain the following form of the general solution of equation (6.2.12)

$$Q(\eta, s) = \frac{1 + \frac{1}{2}s}{\lambda(s)} \tilde{Q}''(s) + P(\eta, s) + \frac{1}{\lambda(s)} \frac{dP(\eta, s)}{d\eta} \quad (6.2.16)$$

where $\tilde{Q}''(s)$ is the Laplace transform of $\tilde{q}''$. The right part of equation (6.2.16) consists of the sum of two terms, which are responsible for the temperature

responses due to the surface heat flux and volumetric heating, respectively. It is possible to represent the function $\tilde{Q}(\eta, s)$ as a product of two functions, one of which depends on the complex variable s only, and the other is $\exp(-\eta/\Delta\eta)$. Hence, the particular solution of equation (6.2.12) can be written as:

$$P(\eta, s) = \Delta\eta^2 \frac{1 + \frac{1}{2}s}{\Delta\eta^2 s^2 + 2\Delta\eta^2 s - 1} \tilde{Q}(\eta, s) \quad (6.2.17)$$

It can be seen that equation (6.2.17) defines two different processes, which are due to electron–phonon and phonon–phonon scattering, characterized by the denominator of the transfer function and numerator, respectively. The relaxation time τ is defined as

$$\tau = \frac{\alpha}{c^2} \quad (6.2.18)$$

where c denotes a speed of sound in a bulk material and corresponds to phonon–phonon scattering [143]. Hence, after taking into account the definition for τ in equation (6.2.18) and dimensionless constant $\Delta\eta$, we obtain the following expression which is simply the relaxation time corresponding to electron–phonon scattering:

$$\tau_{e-ph} = \frac{\kappa}{c} \quad (6.2.19)$$

By substituting (6.2.17) into equation (6.2.13) and finding the inverse Laplace transform, the solution of equation (6.2.7) can be obtained in the integral form. To get the surface temperature, we set $\eta = 0$ and obtain

$$\begin{aligned}
\theta(0, \xi) = & \int_0^\xi \left[\tilde{q}''(\xi^*) + \frac{1}{2} \frac{\partial \tilde{q}''(\xi^*)}{\partial \xi^*} \right] I_0(\xi - \xi^*) e^{-(\xi - \xi^*)} d\xi^* \\
& + \frac{\Delta\eta^2}{4\sqrt{1 + \Delta\eta^2}} \sqrt{\frac{\pi}{a}} \int_0^\xi \left\{ \left(1 + \frac{1}{2} s_1 \right) e^{\frac{B_1^2}{A^*}} \right. \\
& \times \left[\operatorname{erf} \left(\sqrt{A^*} \xi^* + \frac{B_1}{\sqrt{A^*}} \right) - \operatorname{erf} \left(\frac{B_1}{\sqrt{A^*}} \right) \right] \\
& - \left(1 + \frac{1}{2} s_2 \right) e^{\frac{B_2^2}{A^*}} \\
& \times \left[\operatorname{erf} \left(\sqrt{A^*} \xi^* + \frac{B_2}{\sqrt{A^*}} \right) - \operatorname{erf} \left(\frac{B_2}{\sqrt{A^*}} \right) \right] \Big\} \left(\delta(\xi - \xi^*) \right. \\
& \left. - \frac{1}{\Delta\eta} I_0(\xi - \xi^*) e^{-(\xi - \xi^*)} \right) d\xi^*
\end{aligned} \tag{6.2.20}$$

where $I_0(\xi)$ is the modified Bessel function, $\delta(\xi)$ denotes the Dirac delta function, $\operatorname{erf}(\xi)$ stands for the Gauss error function [137], $s_{1,2} = -1 \pm \frac{1}{\Delta\eta} \sqrt{1 + \Delta\eta^2}$, $B_{1,2} = s_{1,2} - 2 \frac{a}{\xi_p^2} \xi_b$, and $A^* = \frac{a}{\xi_p^2}$.

The mathematical model has been developed, based on the assumption that the heating of the electron gas is instantaneous. It shows that the initial heat transfer in thin metallic films occurs by means of electron–phonon interaction, followed by phonon–phonon interaction. The above model predicts that the electron–phonon thermal equilibrium occurs after the laser excitation and defines the electron–phonon relaxation time as the ratio of the penetration depth to the speed of sound in the bulk material, see equation (6.2.19). The model describes the surface temperature response of the thin film induced by an ultrashort laser pulse and takes into account contributions of the transient heat flux (defined by the first

integral in the expression (6.2.20)) and the volumetric heat source (the second integral in the equation (6.2.20)).

6.3 Experimental Setup

The pump-probe transient reflection experimental setup is shown in Fig. 6.1.

Fig. 6.1.

Pump-probe transient reflection: OPO is the Optical Parametric Oscillator; GTP is the Glan–Thompson Polarizer; WLC is the White Light Continuum [164]

The output of Titanium–Sapphire (Legend Eite, Coherent) regenerative amplifier seeded by an oscillator (Micra, Coherent) was used as a pulse laser source: wavelength 800 nm, pulse width 65 fs, pulse repetition rate 1 kHz, average power 3.5 W. 90% of the radiation was converted into the UV (350 nm) by the use of the optical parametric oscillator (Topas, Light Conversion) with the following second- and fourth-harmonic generation and was used as pump pulse. The remaining 10% was used to generate white light continuum in a CaF₂ plate, i.e., the probe pulse [171]. Pump pulses (fluence $\sim 40 \mu\text{J}/\text{cm}^2$) were focused on the surface of a 300- μm -thick gold film deposited on a SiO₂ substrate with a lens of 30-cm focal length and an incidence angle of 10° . Probe pulses with variable time delays relative to pump pulses were used to measure time-resolved transient reflection produced by the pump pulses. The white light continuum was split into

two beams (probe and reference). After reflection from the sample, it was directed into two diode arrays attached to spectrometers (Model 77400, Oriel). According to [59, 172], at early times after 100-fs laser excitation, the normalized reflectivity change $\Delta R/R$ is proportional to the normalized temperature change of the electron gas $\Delta T/T$:

$$\frac{\Delta T}{T} \sim \frac{\Delta R'}{R'} \quad (6.3.1)$$

Thus, the surface reflectivity kinetics were obtained and assigned to the surface temperature variations.

6.4 Experimental Results

Figures 6.2, 6.3, and 6.4 display the normalized reflection changes at 561, 590, and 632 nm, respectively, after 350-nm excitation of the gold films deposited on a quartz substrate (open circles are the experimental data; solid curves denote applied theoretical model).

Fig. 6.2.

Decay kinetics of the reflection change at 561 nm from 300 μm gold plated quartz substrate after the excitation with 350-nm 65-fs laser pulse and theoretical curves for different penetration depths κ

Fig. 6.3.

Decay kinetics of the reflection change at 590 nm from 300 μm gold plated quartz substrate after the excitation with 350-nm 65-fs laser pulse and theoretical curves for penetration depth $\kappa = 3.8$ nm corresponding to electron–phonon relaxation time equal to 1.15 ps

Fig. 6.4.

Decay kinetics of the reflection change at 632 nm from 300 μm gold plated quartz substrate after the excitation with 350-nm 65-fs laser pulse and theoretical curves for penetration depth $\kappa = 3.0$ nm corresponding to electron–phonon relaxation time equal to 0.9 ps

The experimental data in the time range $t = 0 - 5$ ps were also fitted to a multiexponential decay function convoluted with the instrument response function $B(t - t_0)$ centred at t_0 :

$$\Delta R'(t) = \int_{t'=0}^{\infty} \left(\sum_{i=1}^n \Delta R'_i \exp\left(-\frac{t'}{\tau_i}\right) \right) \times B(t - t' - t_0) dt' \quad (6.4.1)$$

where $\Delta R'(t)$ is the differential reflection at time t , $\Delta R'_i$ is the amplitude of component with lifetime τ_i . The full-width-half-maximum (FWHM) of the instrument response function, taken from the pump-probe cross-correlation signal in quartz, was 100 ± 10 fs.

Table 6.1 shows fit results. Rise times at all probe wavelengths were less than 100 fs. Decay times decrease from 1.63 to 0.87 ps by changing the probe wavelengths from 561 to 632 nm.

Probe wavelength, [nm]	561	590	632
Rise time, [fs]	<100	<100	<100
Decay time, [ps]	1.63	1.27	0.87

Table 6.1.

Deconvolution-fit results of rise and decay times of the reflection change in gold films after 350-nm excitation

6.5 Model Analysis and Validation

The first and second integrals in the expression (6.2.20) are denoted by $\theta_f(0, \xi)$ and $\theta_v(0, \xi)$, respectively. Contributions of $\theta_f(0, \xi)$ and $\theta_v(0, \xi)$ are different and depend on ξ_p and $\Delta\eta$. The dependence of the local temperature on $\theta_f(0, \xi)$ has been studied by [12]. However, the analysis of model shown in equation (6.2.20) shows that for the values $\xi_p < 0.02$ and $0.01 < \Delta\eta < 0.15$ (those have been chosen from practical reasons to fulfill conditions of the experiment performed), the contribution of the surface heat flux $\theta_f(0, \xi)$ becomes negligible in comparison with the contribution of $\theta_v(0, \xi)$. Hence, the model given in equation (6.2.20) can be approximated as

$$\theta(0, \xi) \approx \theta_v(0, \xi) \quad (6.5.1)$$

After the laser excitation, the surface temperature is monitored up to 400 ps. To compare the model with experimental data directly, we use the normalized temperature. A maximal temperature change is 350 K as estimated from equation (6.2.9). Hence, we are operating in the linear range of reflectivity versus the temperature [173, 174]. Gold properties and their dependence on temperature are presented in Table 6.2.

Temperature, [K]	300	400	500
Thermal diffusivity ^a α , [m ² /s]	1.27×10^{-4}	1.22×10^{-4}	1.19×10^{-4}
Speed of sound ^b c , [m/s]	3280	3200	3140
Relaxation time ^c τ , [ps]	12	12	12

Table 6.2.

Temperature-dependent bulk properties of gold.

^aReference [175], ^breference [176], ^ccalculated by equation (6.4.18)

The variation of the relaxation time τ is only 2.7% in the temperature range between 300 and 500 K. Hence, the influence of the local surface temperature variations on τ is negligible. Thus, the relaxation time can be taken as a constant equal to 12 ps.

Now, taking into account the value of the relaxation time and the time characteristics of the laser pulse, the dimensionless variable $\xi_p = 4.2 \times 10^{-3}$. Since the heat is dissipated in a surface layer of the gold film and the radiation penetration depth lies within the range 2 to 10 nm or, in the dimensionless form, between 0.03 and 0.1, equation (6.5.1) is valid.

Figure 6.2 displays a comparison of the experimental results at the probe wavelength 561 nm with the mathematical model in section 6.4 under variations of the radiation penetration depth. A good agreement was found between the theoretical model in equation (6.5.1) and experimental results for the penetration depth of 5.0 nm that corresponds to electron–phonon relaxation time $\tau_{e-ph} = 1.52$ ps calculated by equation (6.2.19) and phonon–phonon relaxation time $\tau = 12$ ps.

Figures 6.3 and 6.4 present kinetics of the reflection changes at 590 and 632 nm measured experimentally, together with the data obtained theoretically. Calculations were performed for the radiation penetration depths of 3.8 and 3.0 nm which correspond to electron–phonon relaxation times calculated by (6.2.19) $\tau_{e-ph} = 1.15$ and 0.9 ps, respectively.

6.6 Conclusion

Theoretical and experimental studies were conducted for the heat transfer processes in thin gold films induced by 100-fs laser. The experimental results analysis shows that the occurrence of heat transfer in thin gold films happens via electron–phonon and phonon–phonon interactions. The resulting temperature wave is dissipated in a surface layer of thin films within 800 fs. Electron–phonon relaxation time varies in the range from 1.6 to 0.8 ps with the wavelength $\lambda_p = 560\text{--}630$ nm. The phonon–phonon relaxation time is equal to 12 ps and dependence on the probe wavelength appears to be weak.

The proposed model is a result of TTM transformation to the hyperbolic heat equation based on the assumptions that the electron gas is heated instantaneously and Cattaneo’s law is applied to the phonon subsystem. The analytical studies of the model were conducted within a semi-infinite domain with both the volumetric and the surface heat flux energy sources and the result agrees well with the experimental data. The view of heat processes proposed here allows us to define the time of electron–phonon relaxation as a ratio of the penetration depth to the speed of sound in the bulk material. In turn, this definition of the electron–phonon relaxation time provides an explanation of experimental results, which show the dependence of the electron–phonon relaxation time on the wavelength, and agrees well with the experimental results.

Chapter 7. Summary, Conclusion and Discussion of Results

7.1 Summary and Discussion of the Results

In this chapter the results obtained during this research are discussed, the ways of harnessing the phenomena studied are proposed and possible future issues on further investigations are highlighted.

7.1.1 Theoretical Model of Heat Transfer

In Chapter 3, the quantum mechanical aspect of the wave dynamics was investigated. First, the Schrödinger equation was studied in terms of possibility to obtain some macroscopic parameters of the particle movement. The result is the splitting of the equation into two parts: one is the classical Hamilton-Jacobi equation describing the action of the particle and another is simply the classical equation of continuity. Hence, it is shown that in this case the equation describes the behavior of the probability density (of finding a particle in some specific point at a given moment) “traveling” in the space with velocity v .

The Navier-Stokes equation was studied for the case of incompressible fluid flow with the use of corresponding de Broglie's wave's phase representation. This approach has led to the Navier-Stokes equation being transformed into the reaction-diffusion equation written in terms of wave function corresponding to the de Broglie's wave of a fluid particle moving with velocity v . This leads to the conclusion that the fluid becomes inviscid as long as the wave packet exists only, because as the wave packet starts to widen, the dissipative processes begin, i.e. diffusion.

This time span of the wave packet existence was estimated using the non-relativistic Schrödinger equation and de Broglie's equation. During this time span a set of particles can be considered as a narrow wave packet. After this time span the wave packet starts to widen and the diffusion processes become dominant and, hence, the flow can no longer be considered as inviscid.

It is shown in this thesis that that the macroscopic equation for the temperature wave (second sound) follows directly from the equation of the wave packet motion. This conclusion was based solely on the principles of quantum theory, unlike the similar Landau's equation, which is applicable only for liquids and gases. In this case, the above conclusion is valid for any kind of medium including solid-state bodies.

7.1.2 Apparent Thermal Viscosity and Diffusivity and Steady State Heat Transfer

This chapter discusses the phenomena happening in the liquid at ultra-short times. Using method of Laplace transforms, it was shown that the apparent kinematic viscosity increase (up to 1.4 times) happens in a viscous liquid during relaxation time τ_v due to viscous effects. Analogous result for the apparent thermal diffusivity is obtained. However, under certain conditions and with a certain fluid (with Prandtl number approximately 20) it is possible to find such a regime with relatively big distance between two “peaks” of apparent viscosity and thermal diffusivity, so that during the time between these two points a liquid can be considered as a “superfluid”: apparent thermal diffusivity is on its peak when the apparent viscosity is small.

The study of the heat transfer within a nanoscale spatial domain was conducted in order to obtain the expression for the thermal conductivity as a function of a spatial variable. It was shown that the effective thermal conductivity becomes less than the bulk value when the size of the transporting medium is less than the characteristic length of this medium. Also, when the size is much larger than the characteristic length, the effective thermal conductivity becomes the same as the bulk thermal conductivity. Hence, there is a critical point at which the size of the transporting medium is equal to the characteristic length, beyond which the value of the thermal conductivity decreases.

It appeared that the size-dependent thermal conductivity has the maximum, which occurs when the size of the transporting medium becomes 1.53 times larger than the characteristic length of this medium. The maximal value of the thermal conductivity is shown to be about 40 percent larger than its bulk value. The theoretical estimates appear to be in good agreement with the experimental results. The study shows that the heat transport properties of the medium characterized by the heat transfer coefficient are changed with size of the domain and when the size becomes comparable with the characteristic length of the medium under consideration, the heat transfer coefficient starts to increase and then, when the size continues to tend to zero, the heat transfer coefficient achieves its finite upper limit instead of infinity predicted by the classical Fourier law.

7.1.3 Apparent Sources

The formula for the volumetric heat source or sink term was obtained for the diffusion equation, then the apparent source/sink term was added and the solution was deduced from the previous one. The expression showing the influence of volumetric and apparent sources on the respective temperature was presented. The solution obtained for the specific form of laser excitation (Gaussian pulse) was extrapolated for the case of viscous fluid flow to show the influence of volumetric and apparent source on the velocity of the fluid.

7.1.4 Model Validation

The model validation was conducted by doing an experiment, which involved inducing heat transfer processes on thin gold films with an ultrashort pulse (100 fs) of laser excitation. The two-temperature model (TTM), which can be expressed in terms of two coupled nonlinear partial differential equations for one-dimensional problem, is employed here to study the results of the experiment. This model describes the surface temperature response of the thin film induced by an ultrashort laser pulse and takes into account contributions of the transient heat flux and the volumetric heat source. The analytical studies of the model were conducted within a semi-infinite domain with both the volumetric and the surface heat flux energy sources.

The analysis of the data obtained shows that the heat transfer in thin gold films occurs via electron–phonon and phonon–phonon interactions. The resulting temperature wave appears to be dissipated in a surface layer of thin film within 800 fs. Observed decay times of 1.6 to 0.8 ps correspond to the electron–phonon relaxation with the wavelength $\lambda_p = 560\text{--}630$ nm, and 12 ps corresponds to the phonon–phonon relaxation. Moreover, the electron–phonon relaxation time varies versus the probe wavelength; at the same time, the phonon–phonon relaxation time shows a weak dependence on the probe wavelength. The view of heat processes proposed here allows us to define the time of electron–phonon relaxation as a ratio of the penetration depth to the speed of sound in the bulk material.

7.1.5 Proposed Ways of Harnessing the Phenomena

The phenomena discussed and studied above can be harnessed in order to obtain the desired conditions in micro and nanofabrication:

- it can be proposed to seek ways to lengthen the time lag of the medium response with applying short laser pulses to the flow as it was shown that during lag time the wave packet still remains narrow and the fluid can be considered as inviscid. That will keep a layer of particles always excited during this time span and, hence, remaining a structure of narrow wave packets.
- the phenomena of apparent viscosity and thermal diffusivity increase can be used in order to find such a liquid (as it was shown the best candidates are with Prandtl number approximately 20) with maximum distance between two “peaks” (apparent viscosity and thermal diffusivity maximums), so that the apparent thermal diffusivity would be at its maximum and apparent viscosity – close to minimum, hence, the liquid could be considered as a “superfluid-like”. This regime could be maintained with a series of laser excitations as well.
- apparent sources can be used in terms of lengthening the time span of the “superfluid” regime. Finding the regime of laser excitations applied on the flow to make the fluid go faster could be a challenge in order to produce fast flow, which could transport energy quickly.

7.2 Conclusion

7.2.1 Conclusion and Results

The resulting model may be applied, in general, to all circumstances, where heat transfer occurs at nano-scales or during ultra-fast processes. The goal of this study was to achieve understanding of processes at nanoscale level and develop the adequate mathematical model.

The results of this thesis are:

- the fundamental understanding of the physical processes at nano-scales and ultra-fast transport processes acquired;
- experimental data obtained validating the mathematical model. These data can be used in improvement of cooling of electronic devices, enhancement of efficiency of solar cells, etc.
- the rigorous derivation of phenomenological phase-lagging formulation of ultra-fast transport processes from the quantum theory, as other models are proved to be inconsistent in the general case.

- the apparent source/sink term is added to the diffusion equation and solution for the specific laser pulse (Gaussian) is obtained for cases of temperature and fluid velocity.
- it is shown that the phenomenon of second sound exists not only in gases and in liquids, but also in solids.
- the behaviour of apparent thermal diffusivity and viscosity during short time spans after medium excitation is studied.
- the steady state heat transfer is investigated in order to express it as a function of spatial variable.
- the experimental results of ultrafast laser excitation in thin gold films obtained, which agree with theoretical predictions.

Possible applications and further areas of this study may result in:

- Development of thermal sensing mechanism and technology for contact detection;
- Thermal analysis and management in heat transfer across head-disk interface (HDI) of heat assisted magnetic recording (HAMR);
- Thermal actuation analysis for flying height (FH) control.

- Development of a cooling system using a system of nanochannels.
- Other energy super-transport phenomena at nano-scale channels.

7.2.2 Possible Future Issues and Investigations

Possible areas of further study here could be:

- investigation of related relativistic effects, if there are any, in the fluid flow in the nanochannel;
- further studies of apparent thermal diffusivity and viscosity increase, the exact way on how to lengthen the span of time between two “peaks” in order to get a “superfluid” flow;
- apparent source/sink terms and their influence on the fluid flow motion and possible ways of lengthening the lag time, maybe with the use of multiple apparent sources/sinks.
- The phenomenon of high-temperature “superfluidity”, which is one of the main focuses of this project, if harnessed, can be used for enabling super-transport of energy, that is, transporting huge energy packets within ultra-short spans of time. This can lead to another major project that may result into developing a cooling system for micro-electronic and nano-devices, as well as quantum computers, which has no analogs in the world.

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