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Nanoscale heat transport

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New physical phenomena connected with heat transport in structures with dimensions comparable with characteristic lengths of energy carriers are briefly reviewed. Problems with basic physical understanding of mechanisms responsible for energy transport in such structures are considered. In particular, the role of boundaries is discussed. Thermal properties of a few structures which dimensions influence heat transport, namely: superlattices and multilayered systems, nanoporous materials and nanotubes are analyzed. Problem of hot spots in electronic devices is also mentioned. The last part of the paper is devoted to methods of experimental investigation of thermal properties of nanostructures. Capabilities and shortcomings of two relatively new experimental techniques: picosecond reflectance thermometry and scanning thermal microscopy have been discussed.

Key words: *heat transport; nanostructure; nanoscale*

1. Introduction

Progress in technology created possibilities for fabrication of structures with dimensions of a few nanometers used in practical applications. Quantum dots, quantum wires and nanotubes became parts of electronic and optoelectronic devices [1, 2]. Thin films are also commonly used in various fields of technology. In many cases, thermal properties of nanoscale structures become very important. In construction of electronic devices the problem of heat abstraction form active regions must be solved. Thus, materials with possibly high thermal conductivity are demanded. On the other hand, there is also need for materials and structures with a low thermal conductivity which can play a role of thermal barriers. This is why knowledge of thermal properties of nanomaterials is important.

Thermal conductivity of thin dielectric films decreases with decreasing film thickness for films thinner than about 10 μm [3]. For films with thicknesses of about 100 nm, the thermal conductivity is typically 3–4 orders of magnitude lower than that

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of bulk material. This fact can be explained in the frame of a classical theory of thermal conductivity of solids as the result of growing concentration of defects in the crystalline structure of thin films. A lower limit of the thermal conductivity defines its value for an amorphous sample [4]. On the contrary, analysis of heat transport in structures of nanometer size cannot base on the classical approach and entirely new physical effects must be taken into account. A question arises when it is justified to use the term "nanoscale". The mean free path of electrons in metals is about 10 nm, and that of phonons in insulators and semiconductors is in the range from 10 nm to 100 nm at room temperature. The wavelengths are about (0.1–1) nm and (1–5) nm, respectively [5]. "Nanoscale effects" occur when the heat transport takes place in a structure of dimensions comparable with characteristic lengths of energy carriers. The next effect typical of the nanoscale is a confinement of dimensions in which heat transport can take place. In the other cases, the classical model of heat transport is usually adequate for interpretation of observed phenomena. Thus, there is no necessity for distinguishing such phenomena as nanoscale effects.

2. Theoretical models

The description of heat transport in a macroscale bases on Fourier's law which relates the heat flux $\vec{j}_Q$ to the temperature gradient $\vec{\nabla}T$

$$\vec{j}_Q = -\kappa\vec{\nabla}T \tag{1}$$

where κ is the thermal conductivity. The form of the above equation testifies diffuse nature of heat transport. It is based on the assumption that energy transfer form warmer to colder regions of a medium is the result of random processes which lead to establishing new equilibrium in a system. In crystalline insulators, this process is connected with chaotic movement of phonons which are scattered by other phonons and structure imperfections. But it is also assumed that the medium can be divided into elementary volumes (subsystems), each consisting of a large number of particles which are in equilibrium. This local equilibrium is described by a local temperature related to the average energy of the subsystem of particles. Thus heat transport is a non-equilibrium process but energy is transferred between subsystems being in equilibrium, and heat flows on distances much longer than the mean free path of energy carriers. In the nanoscale, the mean free path of energy carriers is comparable with the size of a structure in which energy is transported, and the carriers interact mainly with boundaries of the structure. It is not possible to distinguish subsystems containing large numbers of particles in equilibrium. The question of temperature definition arises. In the classical approach, the energy is averaged over a large number of particles which causes that momentary energy fluctuations are insignificant. In the case of a subsystem comprising a few particles or even a single atom, such an averaging makes no sense. A possible solution is to replace the averaging over a large number of particles (space averaging) by the averaging over time. The next problem is connected with the method of description of particle motion. In the classical approach, the mean kinetic energy of a particle is connected with temperature through the principle of equipartition of energy. In quantum mechanics, atoms in solids are treated as oscillators with quantized energy. Collective vibrations of atoms are described as phonons, which means that energy is calculated using the Bose–Einstein distribution. The formula connecting the mean energy of a particle with temperature differs from the one in the classical model. As a result, temperatures calculated from the classical model and the quantum model differ. It should also be noted that the phonon approach excludes definition of the temperature for a single atom.

The definition of temperature plays a key role in description of heat transport in the nanoscale. Various definitions of temperature lead to different theoretical models of phenomena connected with the heat transport. An example is the analysis of thermal properties of the diamond–lead boundary [6, 7].

As is mentioned above, in the nanoscale the mean free path of energy carriers is comparable with dimensions of a structure in which phonons propagate. Then the phonon motion is practically ballistic between structure boundaries and heat transport depends mainly on phonon scattering on the boundaries. Additionally, the boundary separates regions for which temperature can be defined and is a place where the temperature can abruptly change. Thermal properties of the boundary can be characterized by its thermal resistance

$$R_Q = \frac{\Delta T}{j_Q} \tag{1}$$

where ΔT is the temperature change at the boundary, and j_Q is the heat flux through it. The definition of the boundary thermal resistance is identical with the classic one. But in the nanoscale, the thermal resistance is used to describe the influence of the layer with the thickness d and effective thermal conductivity κ_{ef} on heat transport and the thermal resistance of such a layer is

$$R_Q = \frac{d}{\kappa_{ef}} \tag{2}$$

The concept of thermal resistances was used e.g., for interpretation of results of measurements of the thermal conductivity of polycrystalline diamond [8]. In the nanoscale, the thermal resistance characterizes the boundary, not the layer. Two basic theoretical models are used to describe behaviour of phonons at the boundary. In the acoustic-mismatch model phonons reflect and refract at the boundary, similarly to light at the boundary of two media with different refractive indices. The reflection and transmission coefficients of phonons depend on acoustic impedances $Z = \rho u$ of neighbouring media, where ρ is the mass density and u is the velocity of phonons (sound velocity) [9, 10]. This model, however, does not explain scattering of phonons at the boundary of two regions of the same material and identical crystallographic structures but with different orientations in space. According to the acoustic-mismatch model, such an interface "does not exist" for phonons. Scattering at such a boundary can be explained in the frame of the diffuse-mismatch model; phonons at the boundary "forget" where they came from and they are randomly scattered [10]. In this model, each and every interface scatters phonons. Both models are helpful in interpretating experimental data but they do not describe real physical processes occurring at the interface. Thus they should be treated as a simple approximation.

Due to lack of acknowledged theoretical models, analyses of heat transport processes in nanostructures are based on numerical simulations with two approaches used. In the former one, the Boltzmann transport equation is solved numerically [11]. The Boltzmann transport equation is a fundamental equation of microscopic theory of irreversible processes. It allows taking into account quantum effects but requires understanding of phonon processes accompanying the heat transport. The other approach is based on integration of Newton's equation of motion for a group of interacting atoms. The interaction is described through an interatomic potential which is usually empirical [12]. This method is called molecular-dynamics simulations. Molecular-dynamics simulations are entirely classical and do not describe quantum effects.

Models of heat transport in nanostructures are in the initial stage of development. This development is stimulated by very interesting experimental investigations which show that the heat transport in such structures is connected with new phenomena, not observed in micro- or macroscale.

3. Experimental results

The first nanostructures in which interesting effects connected with heat transport were observed are superlattices and multilayers. Both structures contain alternating layers of different materials. In superlattices, the layers are single crystals (with exception of amorphous superlattices [13]), while multilayers are built from polycrystalline or amorphous films. From the point of view of thermal properties, both structures can be divided into two groups: structures in which the mean free path of energy carriers λ is longer than thicknesses of the films d ($\lambda > d$), and structures in which $\lambda < d$. In the former case, thermal properties in the direction perpendicular to the film planes are practically independent of thermal properties of the films and are defined by the properties of film interfaces. In the latter case, thermal properties of the system depend either on the properties of the films or the properties of interfaces. From the point of view of this paper, the case $\lambda > d$ is more interesting. In multilayers, heat transport in the direction perpendicular to the films is influenced by strongly defective regions at layer interfaces. These regions have very high thermal resistances resulting in very low effective thermal conductivities of the multilayers in this direction. The effective thermal conductivity of so called nanolaminates (multilayers containing a few nanometers thick films) is, for example, of the order of 0.1 $W \cdot m^{-1} \cdot K^{-1}$ [14]. Nanolaminates could be applied as thermal barriers protecting machine parts against high temperatures. But the problem with their stability at high temperatures must be solved.

As has been mentioned above, superlattices contain single-crystalline films with sharp interfaces. The characteristic dimension for a superlattice is its space period D. If the condition $\lambda < D$ is fulfilled, the thermal resistance of the superlattice can be calculated as a sum of thermal resistances of separate films and interfaces. The periodicity of the structure does not influence the heat transport. More interesting is the case when $\lambda > D$. Phonons "see" the periodicity of the structure which results in change of phonon dispersive curves and appearance of a band structure in their energy spectrum [15]. The consequence of the change of the phonon dispersive curves is a considerable decrease of the group velocity of phonons [16] and appearance of new scattering processes [17]. Both effects lead to a reduction of the mean free path of phonons and decrease of the thermal conductivity of the structure. In the majority of experimental investigations of thermal properties of superlattices, the thermal conductivity in the direction perpendicular to film planes decreases with the decreasing lattice period D (e.g., [18–20]). However, experimental results show that such a behaviour is not a universal rule [21]. There is also unsolved problem of existence of a lower limit of the thermal conductivity of superlattices. According to [22], this limit is defined by the thermal conductivity of an alloy with the same composition as the superlattice but other researchers claim that such a limit does not exist [19, 23].

Modern electronics is interested in insulators with low relative permittivities. SiO_2, commonly used in electronic device technology, has the relative permittivity $\varepsilon \approx 3.9$. From this point of view, very promising are nanoporous materials. This technology allows obtaining films with relative permittivities $\varepsilon < 2$ [24]. Sizes of pores are of the order of a few nanometers. Investigation of thermal properties of these materials reveals that the thermal conductivity decreases with increasing density of pores, e.g. threefold decrease of the density of porous SiO_2 (Xerogel) is connected with a drop of its thermal conductivity by one order of magnitude [25]. Similar results were obtained for $HSiO_{1.5}$ [26]. The attempt of interpretation of the experimental data in the frame of the classical theory of composite medium [27] did not give satisfactory results.

Very interesting results were obtained in investigations of heat transport in nanotubes. Nanotubes and nanowires are one-dimensional structures. Based on theoretical considerations for carbon nanotubes, their thermal conductivity along the axis can be as high as 6000 $W \cdot m^{-1} \cdot K^{-1}$ at room temperature [28]. This value is almost three times higher than the thermal conductivity of diamond (2200 $W \cdot m^{-1} \cdot K^{-1}$). Such a high thermal conductivity can be qualitatively explained by waveguiding of phonons along the nanotube resulting in a very long mean free path of phonons. Similar considerations carried out for nanowires lead to the conclusion that the thermal conductivity of these structures is considerably lower than that of bulk material [29, 30]. Experimental investigations of thermal properties of carbon nanotubes confirm the theoretical predictions. The measured thermal conductivity of a single-wall carbon nanotube is in the range from 1750 $W\cdot m^{-1}\cdot K^{-1}$ to 5800 $W\cdot m^{-1}\cdot K^{-1}$ [31]. The thermal conductivity of multi-wall nanotubes is lower and does not exceed 1000 $W\cdot m^{-1}\cdot K^{-1}$ [32]. It is interesting to note that such thermal conductivity is characteristic of carbon nanotubes, e.g. the thermal conductivity of BN is much lower [33]. Quantum character of heat transport in nanotubes leads to appearance of an upper limit of heat conductivity. Thermal conductance is quantized, and the quantum of phonon conductance g_0 is [34, 35]

$$g_0 = \frac{\pi^2 k_B^2}{3h} T \tag{1}$$

Nanostructures cause not only changes in phonon dispersive curves but they also modify mechanisms of energy transfer between electrons and phonons, e.g. in MIS transistors the most efficient energy transfer from electrons to phonons takes place in a small volume near the drain [36]. "Hot spots" appear in the structure. Moreover, in polar semiconductors energy is mainly transferred to optical phonons with very small wave vectors. These phonons have very low group velocities, thus heat abstraction from "hot spots" is slow. A possible solution of this problem is to create additional structures which will convert "slow" optical phonons into "fast" acoustic phonons [37].

This review of effects connected with heat transport in nanostructures has only been limited to a few selected examples, however, it becomes clear that to understand processes responsible for energy transport in such structures a lot of theoretical and experimental investigations should be done.

4. Measuring methods

Only a few experimental methods have been reported to investigate heat transport in nanostructures. The first possibility is to create special nanostructures for this purpose. The example is a sophisticated structure produced for measurement of the thermal conductivity of a single nanotube [38]. A few micrometers long carbon multi-wall nanotube with the diameter of 14 nm was supported by two islands. On the top of the islands platinum heaters and thermometers were fabricated by electron beam lithography. The nanotube thermally bridged these islands. Such methods can be used in fundamental investigations of selected objects but each structure is produced to carry out a single investigation.

Probably the most universal method is the scanning thermal microscopy (SThM). The method was proposed by Williams and Wickramasinghe in 1986 [39]. Its operation principle is based on determination of temperature distribution at a sample surface by a sharp temperature-sensing tip. Present-day equipment for the SThM is typically an optional module of atomic force microscopes. Scanning thermal microscopes can operate in both steady-state and time-variable modes [40]. In the former case, the microscope allows imaging of temperature distribution connected with dc heat sources in the sample. This mode can also be used for measurement of local thermal conductivity of a sample [41] but modulation methods, based on ac temperature field generated in the sample, offer a better spatial resolution and allow using lock-in detection to improve the signal-to-noise ratio [42].

The SThM provides a possibility for determination of local thermal conductivity in the range (0.1–320) $W\cdot m^{-1}\cdot K^{-1}$ with 0.2 $W\cdot m^{-1}\cdot K^{-1}$ resolution [43, 44]. Spatial resolution is limited by two factors – mean free paths of energy carriers and the size of a tip. The former factor determines the resolution of measurements carried out in air. It is in the range of (0.3–0.5) μm [45], being insufficient for examination of nanostructures. In liquids, the main free path is (0.1–10) nm and it is of the order of interatomic distances [43], so the resolution is limited by the size of the tip. At present, the resolution higher than 50 nm can be achieved. The SThM is a very promising technique but the analysis of experimental data is not a trivial task. Heat transport between the tip and the sample depends on a few mechanisms, whose contributions to the measured signal are difficult to evaluate [46]. This is why understanding of physical processes accompanying the SThM measurements is a basic condition of a proper interpretation of experimental data. A more detailed description of this technique can be found elsewhere [43, 47].

The method which can be used to investigate thermal properties of thin films is the picosecond reflectance thermometry (PRT) [48], being a modification of the classical flash method based on analysis of temperature response of a sample heated by a short light pulse [49]. Use of picosecond laser pulse for both generation of temperature disturbance and detection of temperature changes allows investigation of films with thicknesses of about a few tens of microns. PRT was used in investigations of thin metal films [50, 51], thermal resistance of film boundaries [52] or superlattices [53]. A theoretical model of PRT was put forward by Cahill [54]. Analysis of experimental data is obscured by the fact that at times as short as picoseconds, the equilibrium between various parts of the system (e.g. electrons and phonons) might not be reached. Thus the problem of the definition of temperature arises again.

The above examples do not exhaust possibilities of investigation of thermal properties of nanostructures. It should be underlined, however, that independently of the chosen method, a special attention should be paid to the analysis of experimental data. As results from the previous section, processes responsible for heat transport in the nanoscale are rather fragmentarily recognized, and theoretical models describing experimental data are quite complex.

5. Conclusions

Heat transport in the nanoscale is a very interesting field for both theoretical and experimental investigation. The knowledge of phenomena influencing energy transfer in structures with sizes from a few to a few tens of nanometers is far from being complete. There is a lot of work to be done for understanding physical foundations of experimentally observed effects. However, phenomena of heat transport in nanostructures are important not only for physicists. As nanostructures are used in technology, these problems are also meaningful in practice. New effects observed in nanotubes and multilayers can be utilized for fabrication of new materials with demanded thermal properties. More information about nanoscale heat transport can be found in review papers [47, 55] and in the book [56].

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