International Journal of Heat and Mass Transfer 52 (2009) 4302–4307

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

Effective thermal parameters of layered films: An application to pulsed photothermal techniques

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ARTICLE INFO

Article history:
Received 25 September 2008
Accepted 31 March 2009
Available online 22 May 2009

Keywords:
Laser pulse
Heat diffusion
Thermal parameters
Layered-system

ABSTRACT

Pulsed photothermal techniques provide useful methods based on linear relations between measurable quantities to obtain the thermal diffusivity and thermal conductivity of homogeneous materials. In this work, the effective thermal parameters of two-layered films are defined starting from an homogeneous layer which at the surfaces, produces the same temperature fluctuations and the same photothermal signal that the composite heated by a fast pulse-laser. Our theoretical model predicts that the effective thermal parameters of the layered system can only be calculated in the limit when the laser pulse duration is smaller tan the characteristic time of each layer, respectively. The temperature distribution is calculated in each layer by using the Fourier integral and the time-dependent one-dimensional heat diffusion equation with appropriate boundary conditions according to the experimental conditions. Within this approximation, we found an analytical expression for both, the effective thermal diffusivity and thermal conductivity which depend significantly on the thickness and the thermal parameters of each film.

1. Introduction

The measurement of thin film thermal conductivities and thermal diffusivities is a long standing problem in many areas of physics and engineering. As one example, we mention the interest in thermal parameters of dielectric thin film materials used for optical coatings. Another important field of interest is films used in thermal management applications with semiconductor circuits. Such measurements also gained a tremendous interest in connection with the development of thin film diamond-like materials promising excellent thermal properties [1].

The methods used to measure the thermal conductivity are divided in two groups: the steady-state and the nonsteady-state methods. In the first one, the sample is subjected to a constant heat flow and the thermal conductivity (κ) is directly measured after equilibrium has been reached. In the second group, a periodic or transient heat flow is established in the sample and the thermal conductivity is not directly measured yet, but the thermal diffusivity (α). In the case of homogeneous samples, the thermal conductivity is then calculated from the thermal diffusivity.

On the other hand, the determination of effective properties, such as thermal, elastic, dielectric etc., of inhomogeneous systems has long been an important research subject since these effective properties are critical in design and control parameters for successful applications of the inhomogeneous system. Basically, the effective properties are a function of the constituent properties, interfacial characteristics between the different materials of the composite, volume fractions and microstructure of the system. Here, microstructure means the shape, size orientation and spatial distribution of the inclusions.

In particular, layered films are used in numerous devices such as micro-electronic elements, thin-film superconductors, reactor walls and numerous other applications. In recent years, engineers and physicists have paid more attention to the technologies related with thin films because micro-electromechanical systems are required in various applications. At the same time, due to the advanced of short-pulse laser technologies and their application to modern microfabrication technologies, ultrashort pulse heating of thin film structures has been developed rapidly. For example, transient thermoreflectance, photothermal deflection, photoacoustic and optical heating and electrical thermal sensing methods have been well developed to measure thermal properties of thin films [2]. At present, the microfabricated devices method [3] and the 3ω method [4] are the main measurement techniques to obtain thermophysical properties of materials in such low dimensions and small scales.

In recent years, there has been some interest in the thermal characterization of two-layer systems using photothermal techniques. From the analogy between thermal and electrical resistances used in heat transfer problems, Manzanares et al. [5] calculated the effective thermal diffusivity of the two-layer system as a function of the filling fraction of the composite system, the

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doi:10.1016/j.ijheatmasstransfer.2009.03.068

Nomenclature

c	heat capacity per unit mass, J/Kg K	*Greek*	
d	thickness of one layer, m	α	thermal diffusivity, m^2/s
d_i	thickness of the i layer, m	κ	thermal conductivity, W/m K
I_0	intensity of incident laser radiation, J/m^2t	ρ	density, Kg/m^3
$Q(z,t)$	heat flux W/m^2	τ	laser pulse time duration, s
t	time, s	ω	frequency, s^{-1}
$T(z,t)$	temperature distribution, K		
z	coordinate direction, m		

thermal diffusivities and the ratio of the thermal conductivities of each layer. More recently, the effective thermal diffusivity and conductivity of layered systems have been analyzed solving the one-dimensional heat diffusion equation for each layer with appropriate boundary conditions at the interface and at the surface of the composite according the photothermal experiments. Then, the effective parameters of the inhomogeneous system were calculated from an homogeneous material, with the same boundary conditions at the surface, which produce the same physical response under an external perturbation in the detector device [6,7]. These effective parameters, of course, depend strongly on the experimental set up and how they are measured. For example in photoacoustic experiments, depending on the position of the microphone there are two possible detection configuration: the microphone is at the front of the illuminated surface (close cell configuration) or it is at the rear opposite surface (open cell configuration). Thus, according to this consideration, the value of the effective parameters of the composite are not unique but depend on the experimental configuration [7].

In this paper, we present a theoretical investigation of transient heat transport in one and two layers for different laser pulse duration time τ. It is important to emphasize that in the theoretical models and photothermal experiments mentioned in Refs. [5–7], the incident laser radiation is periodically modulated by a chopper for different frequencies.

In order to obtain the effective thermal parameters, the heat diffusion equation is solved assuming that the sample is optically opaque, i.e., all the incident light is absorbed at the surface of the sample. The effective thermal parameters can be determinated by using the same boundary conditions for both, the one and two-layer samples according to the photothermal experiment and forcing the temperature fluctuation of the one (homogeneous sample) and two-layers be equal at the front and rear surfaces. We show that the effective thermal diffusivity and thermal conductivity obtained from the front-surface illumination and the rear-surface are different, the temperature fluctuations for one and two-layer systems are the same if the laser pulse duration is smaller than the characteristic time of the layered composite. It is important to mention the in this work the effects of the interface thermal resistance and lagging [8] on the heat transport in the layered systems are neglected. These latter effects, the electron–phonon interaction and carrier diffusion influence will be considered elsewhere. Interfacial thermal resistance has been show that is important in layered systems with structures that varies on the length scale of several nanometers heated by a short laser pulse. The thermal conductance of many solid–solid interfaces have been studied experimentally but the range of observed interface properties is much smaller than predicted by simple theory [9–11].

The results presented in this work show that in general an effective thermal parameters for two-layer systems cannot be defined as in photothermal experiments when the heat diffusion in the composite system is created by a periodic light beam.

2. Theoretical model

It is well known that heat transport in solids is carried out by various quasiparticle systems (electrons, holes, phonons, etc.), the interactions between these quasiparticles are such that each of these systems are describes by their own temperature and the physical conditions at the boundaries of the sample may be formulated separately for each quasiparticle system [12]. However, for small effective cooling length of the quasiparticles systems as compared with the sample dimensions and strong interaction between them, the system of quasiparticles can be described by the single particle approximation and the coupled heat-diffusion equations reduce to the usual diffusion equation [13]. For simplification, we shall consider a sample with the form of parallelepiped. On one of the surface ($z = 0$) there is an incident pulsed laser excitation, the other one at $z = d$ is maintained at constant temperature T_0, and the lateral faces are adiabatically isolated. In this geometry the one-dimensional heat diffusion equation is given as

$$\frac{\partial T^2(z,t)}{\partial z^2} = \frac{1}{\alpha}\frac{\partial T(z,t)}{\partial t} \tag{1}$$

where the thermal diffusivity α is given by $\alpha = \kappa/\rho c$, κ the thermal conductivity ρ the density and c the heat capacity of the sample, respectively.

Let us assume that a rectangular laser pulse with an arbitrary duration τ and intensity I_0 is incident on the surface of a layer sample of thickness d, and thermal parameters α and κ. In addition, we consider for simplicity that the one-layer system is optically opaque, i.e., the total laser radiation is completely absorbed on the surface and converted into heat, see Fig. 1(a). Then, the temperature fluctuations $T(z, t)$ in Eq. (1) should be supplemented by boundary conditions at the surface of the sample and some initial conditions. In transient heat transport experiments, the most common mech-

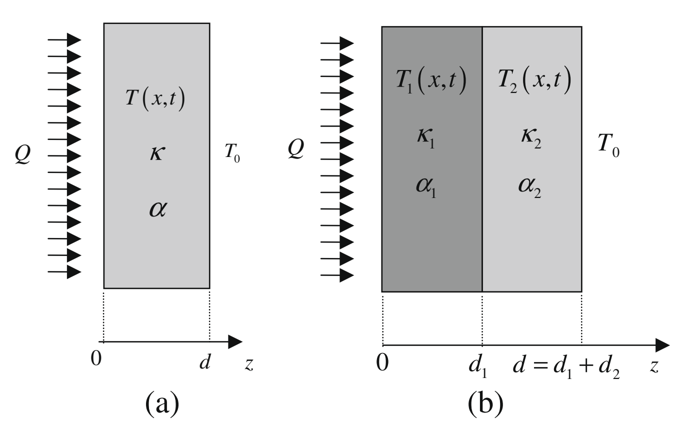

Fig. 1. Geometry for (a) a one-layer system with effective thermal conductivity κ and effective thermal diffusivity α and (b) two-layer system characterized by a thermal conductivity κ_l, and thermal diffusivity α_l for $l = 1,2$.