UNIVERSITI PUTRA MALAYSIA

DEVELOPMENT OF A NEW CONVERGING THERMAL WAVE TECHNIQUE FOR DIFFUSIVITY MEASUREMENT OF HIGH CONDUCTIVITY THIN FOILS

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FS 2008 4

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By

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Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Master of Science

May 2008

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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

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May 2008

Chairman: Professor Mohd Maarof H.A. Moksin, PhD

Faculty: Science

Major problems in measuring thermal diffusivity by using conventional optical technique arise due to difficulties in detecting the thermal signals particularly from high conductivity material in the form of thin foils. Despite the laser beam being tightly focused to form a heating ring on the sample surface, to the point that it could damage the sample, to induce converging thermal waves, the collected signals remain weak. The problems were overcome in this report by using a very large number of optical heating rings instead of a single heating ring in the conventional method. The thermal wave signals at the centre of the ring were generated subsequent to the absorption of the optical pulse beam in the form of a very large number of concentric rings. The signal was found to fit very reasonably well to the theoretical model signal based on very large number of concentric heating rings. The capability of the new design was proven from the measurement of thermal diffusivity of standard thin foils of Al, Cu, Zn, Ni and Brass of thickness in the range of $(0.75-200)$ μm with accuracy within 1%. In the case of Al and Cu foils, its thermal diffusivity was

successfully measured for much thinner foil of thickness down to 0.75 μm and 7 μm as compare to 50 μm and 30 μm respectively in the previous report.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Sarjana Sains

PEMBANGUNAN BARU BAGI TEKNIK GELOMBANG TERMA MENUMPU UNTUK KERAJANG YANG BERKEKONDUKSIAN TERMA YANG TINGGI

Oleh

Mohd. Shahril bin Husin

May 2008

Pengerusi: Professor Mohd Maarof H.A. Moksin, PhD

Fakulti: Sains

Permasalahan besar dalam pengukuran peresapan terma dengan menggunakan teknik optik konvensional adalah timbul daripada kesulitan dalam mengesan isyarat terma terutamanya daripada kerajang nipis yang mempunyai kekonduksian terma yang tinggi. Walaupun menggunakan sinar laser yang terfokus berlebihan untuk membentuk cincin pemanas pada permukaan sampel, sehingga boleh merosakkan, untuk menghasilkan gelombang terma menumpu, isyarat yang dikumpul masih lemah. Masalah di atas dapat diatasi dalam laporan ini dengan menggunakan cincin optik pemanas yang besar bilangannya menggantikan cincin tunggal dalam kaedah konvensional. Isyarat terma di pusat cincin adalah terjana berikutan dari penyerapan alur denyut optik dalam bentuk cincin optik sepusat yang amat besar bilangannya. Isyarat yang tersebut didapati menepati dengan isyarat teori yang berasaskan kepada pemanas cincin sepusat yang amat besar bilangannya. Kemampuan rekabentuk baru ini telah dibuktikan dengan hasil pegukuran peresapan terma sampel kerajang nipis piawai terdiri daripada Al, Cu, Zn, Ni dan Brass dengan kejituan 1% dalam julat tebal $(0.75-200)$ μm. Dalam kes kerajang Al dan Cu, teknik ini berjaya membuat

pengukuran peresapan terma untuk kerajang yang jauh lebih nipis sehingga mencapai 0.75 μm dan 7 μm berbanding masing-masing 50 μm dan 30 μm yang dilaporkan sebelum ini.

ACKNOWLEDGEMENTS

I am extremely thankful to my supervisor and co-supervisor, who have taken lot of efforts in shaping my research work and in framing the thesis. Apart from the academic guidance, his constant encouragement and firmness have helped me a lot in completing my research within a reasonable time frame. I greatly owe him for all his contributions in completing this thesis. The intellectual discussions I had with Prof. Dr. Mohd. Maarof H.A. Moksin and Assc. Prof. Dr. Azmi Zakaria has been an asset to my knowledge. The influence of his criticism in developing my career and personality, both in academic point of view and in personal regard, is so strong that I will never forget them. Right from the beginning, they have always been there with a pleasant smile and moral support, especially during my tough days on research. It is my happiness to extend hearty thanks to them for all their suggestions and help. I also extend my sincere thanks to Department of Physics and the Universiti Putra Malaysia for allowing me use the research grant in order to supply the samples and facilities for the experiment. I would like to acknowledge the financial support from JPA (Jabatan Perkhidmatan Awam). This project of thesis was presented at seminars and won some awards from local and international level for its new invention of technology [Appendices 1.0-1.3]. I am extremely thankful to my lovely wife and son, Suhaini and Shukran for spending some of their precious time with me. I will never forget the happy and wonderful moments they shared with me. I love to thank to my parents Husin Salamat and Siti Salmiah for their unrelenting supports showered on me with their endless love. I am also unforgotten to thank all my friends for theirs help for my survival along my study. Finally, I owe many thanks especially my wife, Suhaini Hassan and my little son, Hafizul Shukran for undoubted supporting me and share every bitter and sweet moments along my journey in finishing my master of degree.

I certify that an Examination Committee has met on 5 Mei 2008 to conduct the final examination of Mohd. Shahril Husin on his Master of Science thesis entitle "Development of a New Converging Thermal Wave Technique for Diffusivity Measurement of High Conductifity Thin Foils" in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulation 1981. The Committee recommends that the student be awarded the Master of Science.

Member of the Examination Committee were as follows:

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Master of Science. The members of the Supervisory Committee were as follows:

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Date: 11 September 2008

DECLARATION

I declare that the thesis is my original work except for quotations and citations which have been duly acknowledgement. I also declare that it has not been previously, and is not concurrently, submitted for any other degree at University Putra Malaysia or at any other institution.

MOHD. SHAHRIL BIN HUSIN

Date: 5 August 2008

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LIST OF ABBREVIATIONS

NDE	Non-Destructive Evaluation
CTW	Converging Thermal Wave
NCTW	New Converging Thermal Wave
PAS	Photoacoustic Spectroscopy
PTD	Photothermal Deflection
TL	Thermal Lens Spectroscopy
AFM	Atomic Force Microscopy

LIST OF SYMBOLS

a Radius

α Thermal diffusivity

α_r Radial thermal diffusivity

α_z Axial thermal diffusivity

c Specific heat

E Laser pulse energy

κ Thermal conductivity

ε Emissivity

h Convective heat transfer coefficient

ρ Mass density

Q Heat pulse energy

Q_0 Heat source intensity

q Heat flux

T_0 Ambient temperature

T Temperature

$t_{1/2}$ Time for a transient signal to reach half maximum

CHAPTER 1

INTRODUCTION

The thermal properties of solids have been intensively investigated since the 19^{th} century, when A. J. Angstrom reported for the first time his experimental results on thermal conductivity measurement of metals [1]. Up to now variety of methods for studying thermal characteristics of mainly homogeneous bulk materials have been developed [2]. In the very beginning they were applied for basic research of massive metals or binary compounds. Nowadays, the development of new thin-film and nano-structured materials for high-tech and other application is a field of vivid interest in contemporary material science. This stimulates also efforts on development of new methods [3-5] suitable for studying of the thermal properties of new advanced materials. However, it is well known that in principle the bulk material properties differ substantially from that of the thin films whereby in the latter the effect of heat waves interference and reflection at the sample interface may dominate beside the difference in its microstructure [6]. Therefore, the existing methods that were proven suitable for bulk material are no longer useful for the case of thin samples.

1.1 Conventional Methods

The flash method is the most acknowledged technique to measure the thermal diffusivity at high temperatures. It is currently considered a standard for thermal diffusivity of solid materials. It was introduced by Parker and co-workers [7] and consists of heating the front face of an opaque slab by a short laser pulse and detecting the temperature evolution at its rear surface. The thermal diffusivity is obtained by measuring the time corresponding to the half maximum of the temperature rise ($t_{1/2}$), which is related to the thermal diffusivity through the expression: $\alpha = 1.137L^2/\pi^2 t_{1/2}$, where L is the sample thickness and α is the thermal diffusivity. This procedure works well under ideal conditions of negligible heat losses and laser pulse duration as compare to temperature history at the rear surface. Even though negligible heat loss requirements can be fulfilled by curve fit to the complete temperature history data of the rear surface, the short pulse duration has its limitation when the sample becomes thinner or its conductivity becomes higher or both.

As mentioned earlier, as the industry is getting developed, the use of thin layers of high thermal conductivity for the thermal management applications is growing. The thermal diffusivity is one of the most pertinent physical properties of material for thermal applications. As such many techniques have been suggested to measure thermal diffusivity of thin slabs, for instance the 3ω [8], the photothermal technique [9], the laser flash technique [10], and the dc-heated bar technique [11]. However, these techniques are limited to samples of low conductivity and samples of thickness 0.5 mm in case of high thermal conductivity. A converging thermal wave technique [12-13] was suggested to overcome the problems related to thin samples of high conductivity materials. The converging thermal wave technique [12-13] was found most suitable for the very thin sample and to be less accurate for thicker sample. Furthermore, it is pertinent to have heating optical beam in the shape of very thin ring to reduce the error in the ring diameter measurement. This requires a highly focus laser source that could exceed the damage threshold of the sample. Otherwise a larger ring diameter has to be employed to the extent that impairs the signal to be collected where signal amplification and data averaging over up to thousands signals are standard procedure. These problems become more acute as many high conductivity materials such as metals are highly reflective and absorb very small proportion of light to induce an appreciable signal to be measured unless a suitable light absorbing coat is applied on the sample surface. The latter may cause some complication for very thin sample as the added coat on the thin layer has transformed the sample to a new double-layered material.

1.2 Objectives

A new version of converging thermal wave technique is presented in this report to overcome the problems described above. The objectives are

1. To develop a new method of generating converging thermal wave by using a very large number of optical heating rings.
2. To calculate the generated thermal wave signal at the center of heating rings.
3. To measure the thermal wave signal at the center of the heating rings.
4. To use the technique to measure thermal diffusivity of selected high conductivity thin foils of various thickness.

CHAPTER 2

LITERATURE REVIEW

Converging thermal wave technique has been one of most powerful method in photothermal technique to measure thermo-physical properties of inhomogeneous materials and thin samples of high thermal conductivity. Thermal wave propagates in axial or radial direction depending on the geometry arrangement of the experiment setup. The thermal diffusivity measurement is also affected by heat losses factor. Different probe will gives different accuracy in measurement due to the heat lost effect. In this chapter the review will be focused on converging thermal wave (CTW) technique that used to measure thermal diffusivity of metallic sample.

2.1 Photothermal Effects

The interaction of electromagnetic radiation with matter causes absorption, emission, and scattering of radiation. Except for emission and scattering, the absorbed electromagnetic energy is converted to heat by various nonradiative processes and induces changes in temperature, pressure, and refractive index of the medium [14-15]. The discovery of the photothermal effect dates back to Bell's discovery of the photoacoustic effect in 1880 [16], but it is after the invention of the laser that the photothermal spectroscopies became popular. In 1964, Gordon et al. found a beam divergence effect from liquid samples that were placed in a gas laser cavity [17]. This phenomenon was correctly interpreted in terms of the "thermal lens" effect produced by heating induced by the Gaussian laser beam. The thermal lens method soon became a standard technique to detect the thermal energy produced by nonradiative transitions. Since then, various types of photothermal methods have been developed and applied to a variety of problems. Today, photothermal spectroscopy is widely used in physics, chemistry, biology, and engineering [18–23].

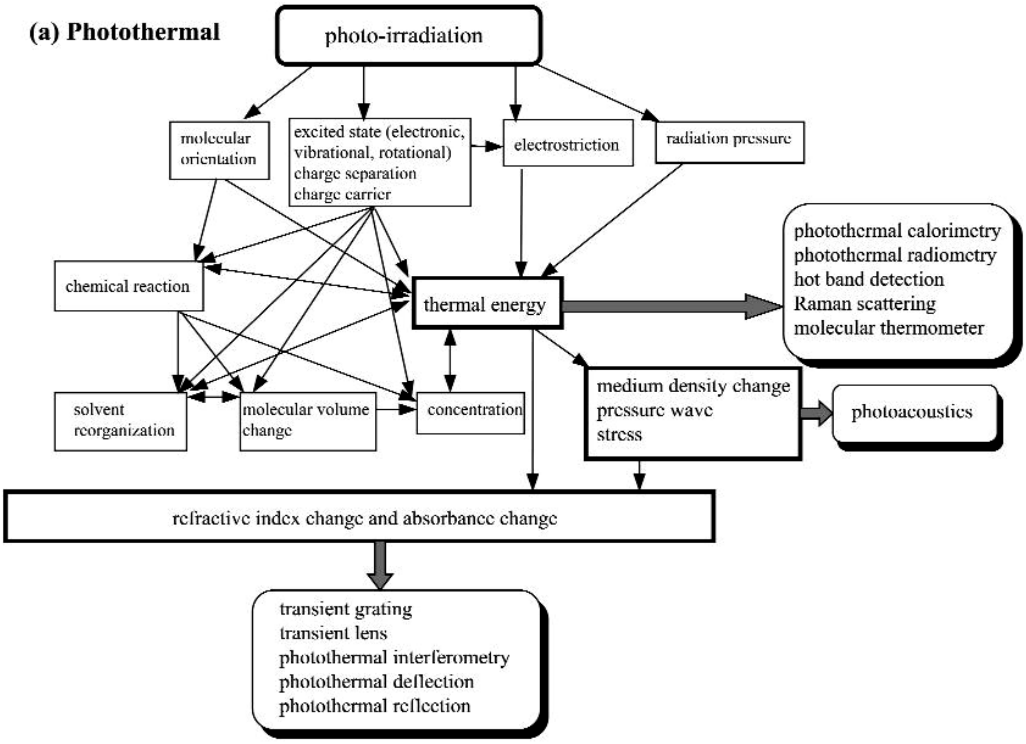
Various changes in the medium can be monitored by photothermal methods in order to quantify the effects of the temperature rise upon radiationless deactivation [18, 20, 22–23]; the temperature rise is measured by laser calorimetry. Furthermore, thermal emission is detected by photothermal radiometry, while reflectivity is detected by transient thermal reflectance.

The photothermal method has a number of merits compared with other methods [18, 20, 22–23]. It is highly sensitive and applicable to different types of materials (gas, liquid, liquid crystal, and solid), transparent and opaque. It can be used in vacuum and in air, and with samples of arbitrary shape. Radiation of any wavelength can be used (radio frequency, microwave, IR, visible, UV, and X-ray, etc.). Photothermal detections are often nondestructive and noncontact methods and can be used to probe optical and thermal local properties in very small areas; these merits are of great value in analytical applications.

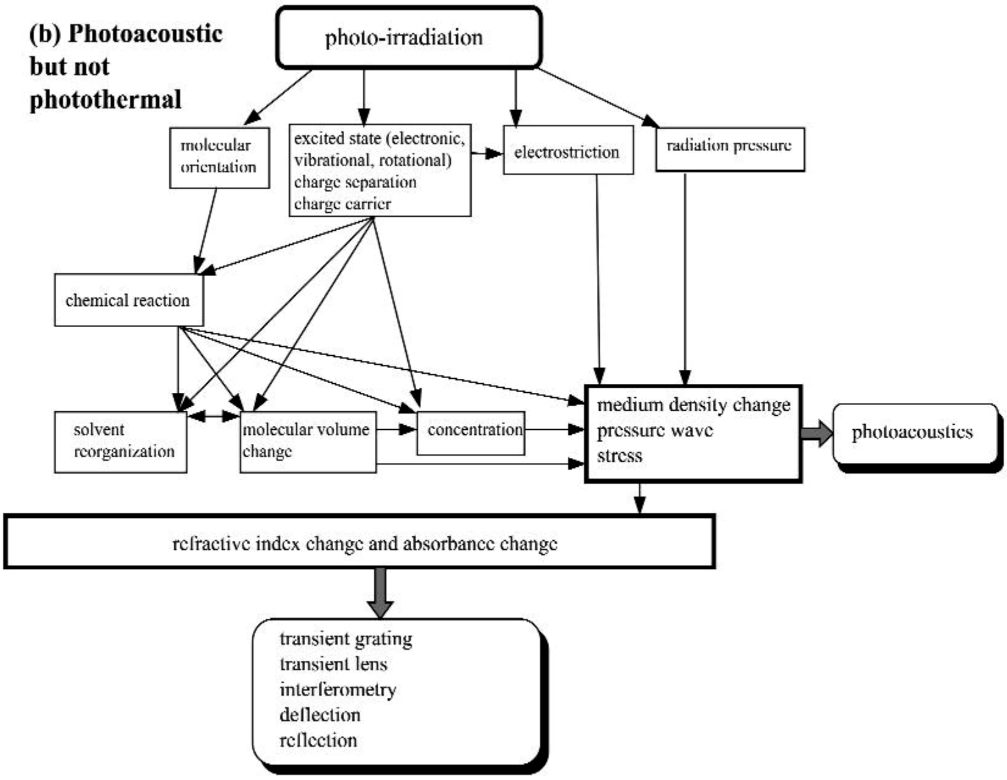
Photothermal techniques are defined as methodologies detecting the heating effect after optical excitation. In as much as various temperature-dependent physical parameters (pressure wave, refractive index, absorbance change, thermal radiation, etc.) are detected, various dynamic processes may be simultaneously monitored. The photo-acoustic (PA) effect is defined by a common effect producing a medium density change, which may be either detected acoustically or optically. The pressure wave generated after photo-excitation contains contributions from various sources, such as radiation pressure, electrostriction, thermo-elastic expansion (by non-radiative transition or thermal energy of chemical reaction), photo-induced volume change, gas evolution, boiling, ablation, and dielectric breakdown. The effect that directly modifies the absorbance or refractive index upon photo-excitation is generally referred as photo-optical effect. The effects listed above are often connected to each other (Figure 2 (a), (b) and (c)). The thermal energy generally creates stress and pressure, and vice versa. Since the energy released by radiationless transitions in condensed phases will eventually flow into translational freedom, the photothermal effect is generally observed after any type of photo-excitation (resonant condition) and is closely related to the energy dynamics in the system. Relation among various photothermal effects [24] are shown in Figure 2.1 (a), (b) and (c). Arrows shows the connection between causes and effects.

Figure 2.1 (a) shows origins and detection methods of photothermal effects, which are caused by the heating effect after photo-irradiation, (b) shows origins and detection methods of acoustic effects induced by photo-irradiation, but not by the photo-thermal effects and (c) shows origins and detection methods of photo-optical effects induced by photo-irradiation, but not by the photothermal effects. Photothermal effects can be probed not only in the bulk phase, but also at surfaces or interfaces as changes in such as intensity, polarization, optical path, and reflection angle of the reflected optical radiation. The effects are the same as those in the bulk.

(a) Photothermal



(b) Photoacoustic but not photothermal



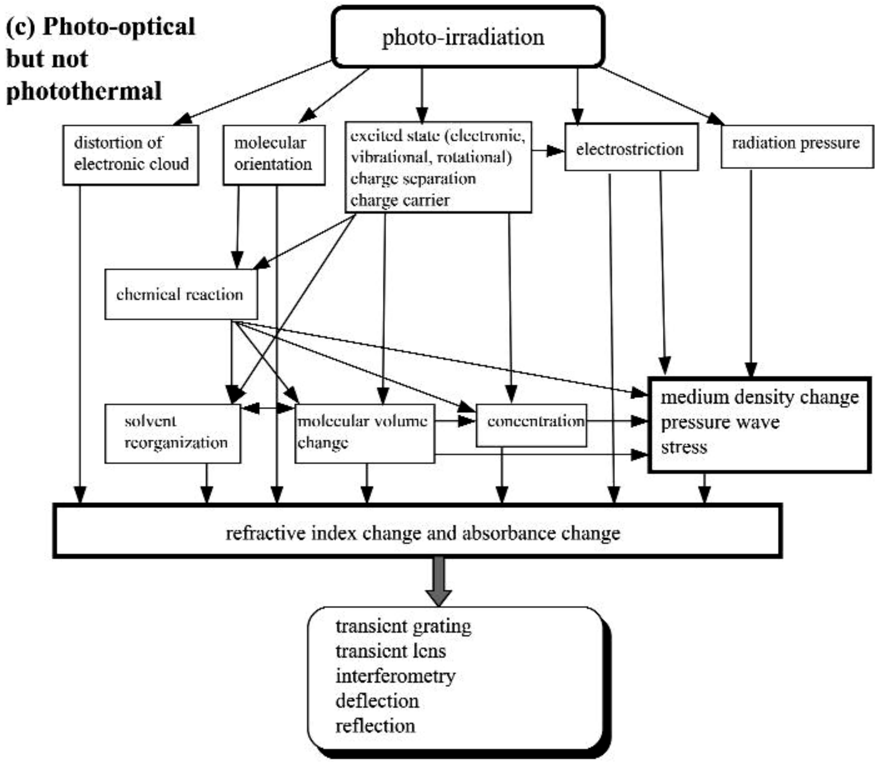


Figure 2.1. (a), (b) and (c). Relation among various photothermal effects. Arrows connect causes and effects [24].

The intensity and reflectivity depend on the surface temperature through the absorption and refractive index changes. Unique to the surface photo-thermal effect is the fact that the reflection angle of reflected radiation changes when the surface is deformed. Relations among various photothermal effects are shown in Figure 2.1.

2.2 Axial Measurement

In 1961, flash method was introduced by Parker [7], the entire front surface is exposed to the energy pulse and the thickness of the specimen is small compared to its diameter, the heat flow is essentially one dimensional in the axial direction. The heat losses are neglected for the small temperature rise. These qualifications of work give rise to a very simple data reduction scheme whereby the theory and experiment need to be matched at only a single point; the time corresponding to one-half of the maximum