The 3-omega Method for Thermal Conductivity Measurements of Thermoelectric Materials

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Abstract

Microelectronic technologies require development of special methods for thermal conductivity measurements of materials in the form of thin layers deposited on metallic or ceramic substrates. The 3-omega method allows accurate measurements of thermal conductivity of bulk materials and layers in the direction perpendicular to the substrate. This paper presents the method and the laboratory arrangement. The procedure was verified on a range of materials: single crystals, polycrystals, and layers, over the thermal conductivity range 0.2 - 140 $Wm^{-1}K^{-1}$.

1. Introduction

One of several techniques, which allow direct measurements of thermal conductivity over a wide range of temperature, is the 3-omega technique, originally developed by Cahill et al. [1-3]. Provided all the requirements are satisfied, this method makes it possible to measure thermal conductivity in the direction parallel λII and perpendicular λ⊥ to the layer surface. An additional advantage is that penetration depth can be controlled, thus enabling characterization of layers, substrates and joints in one measurement. Because of low sensitivity to heat loss by radiation, this method is suitable for materials with high and low thermal conductivities over a wide temperature range.

2. Measuring method

The term 3-omega relates to a broad range of dynamic methods of thermal conductivity measurements. In the case of dielectric specimens it is required that a metallic strip (usually made of gold or nickel) be deposited directly on their surface, this strip playing a double role of a heater and a temperature sensor. The alternating current with an intensity of $I_0 \sin\omega t$, passing across the strip, produces thermal oscillations with a pulse frequency 2ω which results in voltage pulse frequency of 3ω in the measuring strip. The signal carries over the information on thermal properties of the material subjacent to the measuring strip and allows measurements of the amplitude of thermal oscillations, $\Delta T_{2\omega}$.

In this work, we extended the application range of the 3-omega method over conductors of electricity, by depositing on their surface a thin dielectric layer with high thermal conductivity and high resistance to electrical break-throughs (e.g. aluminum nitride AlN or diamond-like carbon DLC), in order to separate the sensor from the layer of the investigated material (Fig.1).

The analysis of temperature changes within the strip deposited on the investigated material is performed with an assumption that the strip thickness is negligible relative to the specimen thickness. It is assumed also that heat flux is two-dimensional, the specimen is semi-infinite and heat conduction in the specimen is isotropic.

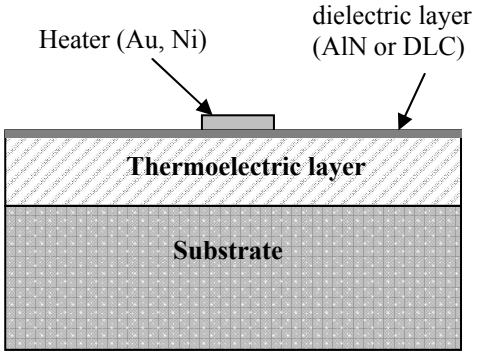

Fig.1 Scheme of specimen cross-section with a sensor (strip of Au or Ni) deposited onto an insulating layer.

In such conditions, the amplitude of temperature oscillations, ΔT, depends on thermal conductivity coefficient λof the material and is expressed as:

$$\Delta \tilde{T}_{2\omega} = \frac{P_l}{\pi\lambda} \cdot \int_0^{\infty} \frac{\sin^2(kb)}{(kb)^2 \cdot (k^2 + q^2)^{1/2}} dk \qquad (1)$$

where *2b* is strip width, P_l power produced per unit length of the strip, $|q^{-1}|$ - penetration depth of the heat wave.

The amplitude of temperature oscillations is calculated from the equation:

$$\Delta T_{2\omega} = \frac{2V_{3\omega}}{IR\alpha} \qquad (2)$$

where: I – current intensity, R – strip resistivity, α - temperature coefficient of resistivity of the strip material, and $V_{3\omega}$ - amplitude of third harmonic component of the measured signal. By solving equation (1) and introducing the amplitude of temperature oscillations, $\Delta T_{2\omega}$, calculated from equation (2), it is possible to calculate thermal conductivity λ.

3. Experimental set-up

The measuring system consisted of an electrical circuit with the Wheatstone bridge, where the measuring element was installed in one of the arms of the bridge, and in other arms there were high-class resistors and potentiometers, having accuracy of more than 0.1%. The circuit was supplied from a generator modulated by a quartz-crystal resonator, which digitally synthesized sinusoidal outputs, with a precisely defined frequency and small contribution of harmonic components (<0.01%). The differential signal from the bridge carried the preextracted signal, $V_{3\omega}$, which was further amplified and recorded by a dual channel, 16-bit recorder with a sampling frequency of 200 kHz. In the second channel, the baseline signal, V_{ω}, was recorded. The

data collected from at least 8 periods were next transmitted to a computer and were numerically processed.

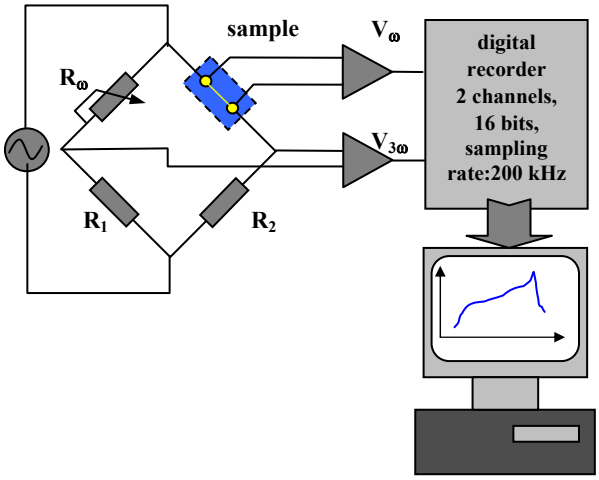

Fig.2 Arrangement for thermal conductivity measurements by a 3-omega method.

The specimen with a sensor strip was mounted on a heating plate in a vacuum chamber, which permitted measurements in nearly adiabatic conditions. The heating plate temperature was controlled with a high precision in the range from 20 to 600 ^{o}C by means of a computer controlled regulator.

4. Preparation of measuring probes

As earlier mentioned, the measurements of thermal conductivity of conductive layers by a 3-omega method require deposition on their surface of a dielectric layer having good thermal conductivity. The most commonly used insulating materials are AlN and DLC fig.3. The insulating layers were sputtered by means of a planar magnetron.

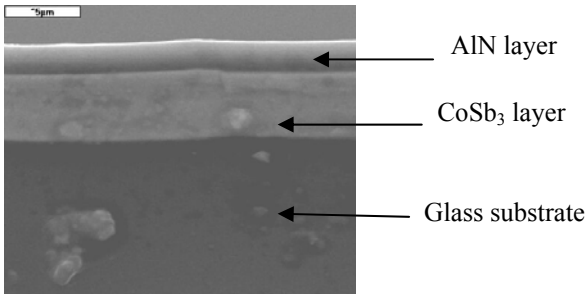

Fig.3 SEM cross-section image of the composite sample.

In this order, the aluminum target with diameter 100 mm was sputtered in reactive gas mixture of nitrogen and argon. The process was conducted slowly, at the pressure of $4 \cdot 10^{-3}$ mbar, for 2.5 to 5 h. Too fast sputtering of Al target lead to an excess of aluminum in the film and lose the dielectric character of the material. Obtained dielectric AlN layers with thickness of about 1µm had good contact with a substrate.

Magnetron sputtering was also used to deposit the measuring probes in the form of thin strips of gold.

The strips were about 60µm thick, and about 4mm long and had circular connections on both ends which enabled connecting of the specimen to the measuring system. Fig.4. shows a ruby single crystal with the deposited gold strips.

The resistance of a gold strip depended on its thickness and varied from 50 to 100 Ω.

Fig.4 Ruby single crystal with the deposited strips of gold.

5. Data analysis

The measuring arrangement was equipped with a specially developed software, 3OMEGA, which enabled processing of the collected data and control of devices and meters. The software was also used for data analysis.

The measured values were plotted in a $\Delta T_{2\omega}$versus f (frequency) co-ordinate system, which enabled finding thermal conductivities of substrates and layers, provided their thickness, d , exceeded the minimum penetration depth of the heat wave, $|q^{-1}|$, equal to:

$$\left|q^{-1}\right| = \sqrt{\frac{\lambda}{2\rho c_p \omega}} \qquad (3)$$

where c_p- specific heat, ρ- density of the material. For illustration, when frequency f is 10 kHz and thermal conductivity of the examined material λ is 10 $Wm^{-1}K^{-1}$, the minimum penetration depth is about 5 µm. Within the data ranges, where linear relationships occur, it is possible to determine the average value of thermal conductivity, independently for the layer and for the substrate, by a linear regression:

$$\Delta T = \frac{-P_1}{2\pi \cdot \lambda} \cdot \ln(4\pi \cdot f) + b \qquad (4)$$

The straight line with a greater slope allows determining of thermal conductivity for the ZrO_2 layer. The value of 2.6 +/-0.1 [$Wm^{-1}K^{-1}$] is higher than ones reported for the zirconia layers and dense sinters. The exact values are given in Table 1. Also, the value determined for the substrate, 45 +/- 9 [$Wm^{-1}K^{-1}$], differs from the reported one of about 72 [$Wm^{-1}K^{-1}$].

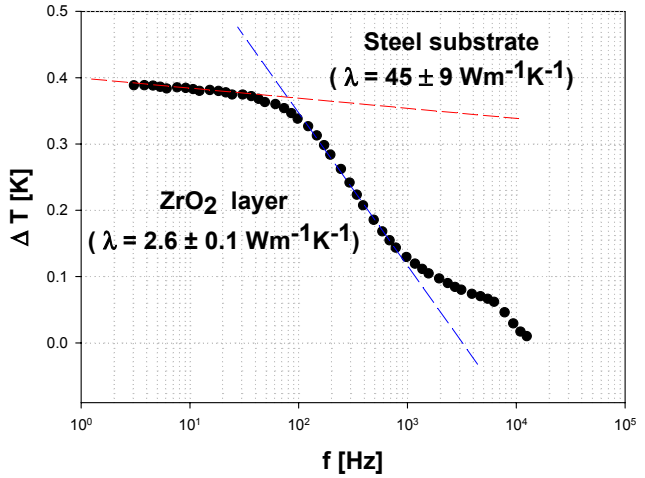

Fig. 5 Amplitude of temperature oscillations of the Au sensor (40 μm x 4mm) versus frequency, for a ZrO_2 layer on an iron substrate.

How can those discrepancies be accounted for? Presumably, the improvement of thermal conductivity of the investigated layer is due to a thin film of DLC, deposited on top of the zirconia layer in order to 'seal' it. Otherwise electrical 'breakthrough' was observed between the Au sensor and the substrate. The 'sealing' material had better thermal conductivity than the zirconia layer itself, and so the measured value might be representative for the ZrO_2 – DLC composite rather than for ZrO_2 alone.

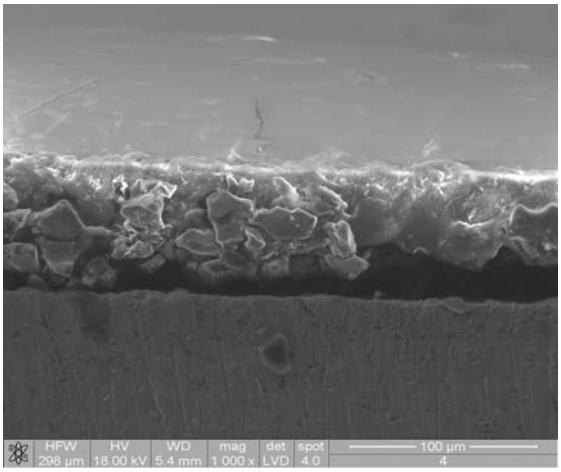

Fig.6 SEM image of the fractured specimen: ZrO_2 layer on iron substrate.

The much lower value of thermal conductivity of the iron substrate is probably related with the specimen morphology. The contact between the zirconia layer and the substrate was not perfect. The measured values would be characteristic of the substrate in the conditions of good adherence. The suspected lack of adherence is confirmed by the microscopic pictures. Fig.6 presents microphotographs of the fractured specimens. It is clearly seen that the zirconia layer is porous and it does not adhere perfectly to the iron substrate. The outer surface of the specimen, due to the deposited DLC film, is quite smooth.

6. Testing of the measuring arrangement

The laboratory arrangement was tested on a range of reference materials in the form of highly pure single crystals and polycrystals. The Au strips, width of 40 μm and length of about 4 mm, were deposited on specimen surface by magnetron sputtering. The measurements by a 3-omega method were made in the generator frequency range 0.1 Hz - 10 kHz and power range 4 mW - 40 mW. Table 1 lists thermal conductivities obtained for different materials tested. They are in good agreement with the reported data.

Table 1. Measured and reported values of thermal conductivity for selected materials at 25ºC.

Material	Reported values [4-7] [$Wm^{-1}K^{-1}$]	Values measured by a 3-omega method [$Wm^{-1}K^{-1}$]	Penetration depth $\|q^{-1}\|$ [μm]
Organic glass (polycarbonate)	**0.19**	**0.21 ± 0.02**	**30 – 90**
Optical glass	**1.1 – 1.5**	**1.05 ± 0.04**	**25 – 240**
ZrO_2	**1.6 – 2.5**	**1.6 ± 0.1**	**25 – 240**
Bi	**7.9 – 8.4**	**7.4 ± 0.7**	**25 – 600**
$CoSb_3$ -film	**10.5**	**10.1±1**	**1.4 – 6.3**
Ruby single crystal	**35 - 40**	**39.5 ± 0.6**	**30 – 1000**
Zn	**112 - 116**	**125 ± 13**	**220 – 2200**
Silicon single crystal	**126 - 148**	**140 ± 14**	**250 – 2500**

7. Summary

The laboratory arrangement for measuring thermal conductivity by a 3-omega method, presented in this work, allows measurements of $\lambda_\perp$ with an accuracy of at least 10% for specimens in the form of thin and thick layers, within the temperature range 20 ºC - 300 ºC. An advantage of this method is that thermal conductivities can be determined in a very wide range, from 0.2 to 140 $Wm^{-1}K^{-1}$, and that it is possible to perform in-depth measurements, over a distance of a few millimeters from the surface. The measured values of thermal conductivity are dependent on specimen morphology.

Acknowledgments

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